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The use of *Carica papaya* seeds as bio coagulant for laundry wastewater treatment

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Abstract

The study aims to analyze the effectiveness of wastewater treatment by using *Carica papaya* seeds as bio coagulants to diminish total suspended solids (TSS) and excess phosphate. This method has some advantages because it does not use chemical materials, is simple to apply, and is safe for the environment and human health. The wastewater samples were taken from an active laundry site in Palembang City, South Sumatra Province of Indonesia. The study found that the best dosage of coagulant was 3 g for the wastewater samples. The pH and BOD parameters showed slight changes after treatment, with pH 7.63–7.19 and 33–27 mg/L values, respectively. The TSS removal efficiency ranged from 9.3 to 15.6%, while the COD and phosphate removals were obtained from 11.7 to 39.3% and 56.3 to 68.4%, respectively. The treated TSS, COD, and phosphate concentrations have met the Indonesian domestic wastewater quality standard and environmental protection organization (EPO) guidelines. In addition, the statistical analysis and *t*-test showed significant differences ($p < 0.05$) for before and after treatment data of all parameters. The ANOVA test showed significant differences ($p < 0.05$) for all parameters among the three treatments. Overall, the study indicated that *C. papaya* seeds are promising materials that are eco-friendly and useful to treat laundry wastewater.

Keywords: *Carica papaya*, Bio coagulant, Laundry wastewater, Suspended solids

Introduction

Environmental degradation through reduction of the qualities of atmosphere and hydrosphere is something that cannot be avoided due to the rapid development and population growth [17, 29]. Flocculation and coagulation are prominent processes that decrease turbidity or suspended solids in the wastewaters [12]. Both processes also enhance the efficacy of advanced treatment methods like filtration, purification, and sedimentation. The coagulants are usually categorized into organic, anorganic, and natural materials. In usual treatment methods, several kinds of coagulants are frequently applied according to the physicochemical characteristics of the pollutants in the wastewater [32]. For instance, aluminum sulfate is widely applied in clean water treatment. However, some studies have reported that applying aluminum sulfate could affect human health [30, 33].

The water treatment technique has recently been oriented to use natural materials for environmental safety and to minimize water treatment costs [8, 10]. Many studies have found natural coagulants such as *Guazuma ulmifolia* [25], *Moringa oleifera* [26], *Alyssum mucilage* [14], *Opuntia ficus-indica* [3], *Azadirachta indica A. Juss.* [35], Moreover, *Carica papaya* [6] could treat wastewater. Several researchers have found that natural coagulants in the *C. papaya* seeds could effectively treat wastewater [2, 20]. Dewi and Rahmayanti [13] found that *C. papaya* seeds could reduce red remazol concentration by over 90% via electrostatic interaction. Hosseini [19] reported that active components of *C. papaya* seeds, like polysaccharides, fiber, and proteins, were helpful in coagulating turbidity in the water. The *C. papaya* seeds also had a high potential to be applied in turbid water clarification [6, 24]. The *C. papaya* seeds could reduce turbidity and total dissolved solids in laboratory wastewater [28]. In Indonesia, the *C. papaya* trees widely grow in all country regions. However, studies about the *C. papaya* seeds for the use of wastewater treatments still need to be made available, especially for laundry wastewater. Thus, this study was carried out using the *C. papaya* seeds for laundry wastewater treatment, where the wastewater samples were taken from an active laundry place in Palembang City, South Sumatra Province of Indonesia, in 2023.

Methods

Wastewater sampling

The wastewater samples were collected from an active laundry place in Palembang City, South Sumatra, Indonesia (2.9761° S, 104.7754° E). The wastewater samples were collected during the dry season (April–June 2023). The wastewater samples were replicated three times during the sampling activity. Wastewater sampling refers to the US Environmental Protection Agency Operating Procedure for Water Quality, sampling, a handbook on the design of sampling schemes, and sampling methods. The wastewater samples were taken around 10–30 cm depth from the top surface of the water. The wastewater samples were collected directly at the starting point of the drainage outlet of the sewer. The wastewater samples were carried to the laboratory for further analysis. This study analyzed several parameters, including pH, BOD, COD, TSS, and phosphate concentrations.

Preparation of *C. papaya* seeds as a bio-coagulant

The coagulant used in this study originated from the seeds of the *C. papaya* species. The *C. papaya* fruit was obtained from the traditional market in Palembang City. The average fruit weight was around 1 kg; we used two ripe fruits with a light, bright orange color. In the laboratory, the papaya seeds were separated from the flesh of the fruit, rinsed, and dried over the sun for 3 days. The dried papaya seeds were then grinded and sieved to obtain papaya seed powder with a size of 0.4 mm. These seed powder were then stored in a desiccator and utilized as a coagulant without farther treatment. The chemical composition of *C. papaya* seeds is shown in Table 1.

Experimental jar experiment

In this study, the jar test experiments (Model JLT-4) were applied in the coagulation process test using the coagulant dosage of 1–3 g for wastewater treatment. A total of 500 ml

Table 1 Characterization of *C. papaya* seeds

Composition	%
Oil	30
Protein	28
Ash	8
Fiber	19
Carbohydrate	25

Source: Kusniawati et al. [23]

of wastewater sample was poured into nine beakers and labeled with the dosage of *C. papaya* coagulant. For an effective coagulation process, the speed of the stirring tool was set at 150 rpm for 3 min and was decreased to 60 rpm for 30 min. Then, the final suspensions were left for 30 min. Lastly, several variables like pH, total suspended solids (TSS), biochemical oxygen demand (BOD), chemical oxygen demand (COD), and phosphate concentration were examined. The jar test trials were carried out at room temperature. The pH of wastewater samples did not show too much change during the process of coagulation, which was around 6.5.

Laboratory analysis

All wastewater samples were analyzed ex situ (in the laboratory) due to the fewer instruments during the field activity. The wastewater parameters, such as pH, TSS, BOD, COD, and phosphate, were determined in the laboratory based on the standard methods for the experimentation of water and wastewater (APHA, 2012). For instance, the COD values were determined using the SMEWW method 5220-D. The BOD values were determined using the BOD5 APHA 5210B method. The TSS values were determined using filtration and gravimetric technique based on the Consult APHA Method 2540 D. The phosphate concentrations were determined using the APHA Method 4500-P. A multi-parameter water quality meter directly measured another parameter, such as pH. All parameters were conducted in triplicates to increase the accuracy of the results. In addition, the removal efficiency (H%) of TSS, COD, and phosphate were determined using Eq. 1 as specified below.

$$\mathrm{H} = \frac{C_0 - C}{C_0} \times 100\% \tag{1}$$

where C_0 and C are the initial and final values of corresponding variables like COD (mg/L), TSS (mg/L), and phosphate (mg/L) in wastewater samples, respectively.

FTIR and SEM analysis

The functional groups that were contained in the coagulant were measured using the FTIR analysis by the infrared spectrophotometer (model Shimadzu). The range of spectrum needed to obtain the functional groups was analyzed at 4000 to 400 cm^{-1}. Moreover, the surface morphology structure of the coagulant was analyzed using the SEM instrument (model Hitachi SU-3500) with × 500 magnification.

Statistical analysis

IBM SPSS Statistics Version 20 was employed for all statistical analyses in this study. A one-way analysis of variance (ANOVA) test was conducted to identify the significant differences in the means of parameters among the three treatments (different dosages of coagulant). Moreover, the independent sample test (*t*-test) was applied to ensure the significant differences between the two means of parameters between pre- and post-treatments.

Results and discussion

pH and BOD changes

The pH level is prominent in the organic and non-organic particle coagulation process. The non-organic colloid particles could be removed significantly by the organic coagulant. The pH value before treatment was 7.63 [15]. If we compared it with the Indonesian domestic wastewater quality standard (pH 6–9), the baseline pH value of the wastewater was still within that range. However, in this study, we also analyzed any possible impacts from the used coagulant to change the pH value of wastewater. Our result showed that the *C. papaya* seed coagulant slightly decreased the pH value of wastewater using 1 g and 2 g coagulant doses (pH 6.4–6.5). Then, it started to increase again after the addition of 3 g of coagulation dose with a pH value of 7.19 (Fig. 1a). A previous study by Amran et al. (2022) that also used *C. papaya* seeds as a natural coagulant showed a similar finding that this coagulant became better pollutant removal at lower pH water. It could also applied in water and wastewater treatments to enhance extreme acidic and alkali pH. Based on the above study, the *C. papaya* seeds could increase the pH of wastewater from 3 to 7. The organic coagulant sustained the pH change when its functional groups obtained protons or split [27]. Therefore, the coagulant from *C. papaya* seeds was appropriate for the laundry wastewater treatment. No extra processes have been needed to improve the pH level after treatment. In addition, after treatment, the BOD values were reduced from 33 to 27 mg/L (Fig. 1b). The pre-treatment BOD was slightly higher than the Indonesian domestic wastewater quality standard (30 mg/L). However, it has met the standard after the treatment process. According to the ANOVA test, the 3 g dosage coagulant BOD value obtained higher significance values ($p < 0.05$) than other treatments. Meanwhile, using a 2 g dosage coagulant did not show any significant differences ($p > 0.05$) with a 1 g dosage coagulant.

Effect of bio coagulant dosage on TSS removal

Studies about the potential of *C. papaya* for total suspended solid (TSS) removal were still rare. In this current study, we obtained TSS removal values that significantly changed when the coagulant dosage was raised from 1 to 3 g (Fig. 2). A study by Abidin et al. [1] found that the natural coagulant could diminish more suspended solids in more turbid water condition. Water with more suspended solids tended to cause more collisions between colloidal and coagulants. This condition would contribute to more opportunities for the particles in water to interact with each other to shape large flocs for the gravity settlement. The highest efficiency of TSS treatment reached 15.6% when adding a coagulant dosage of 3 g. The natural coagulant would generally be negatively charged

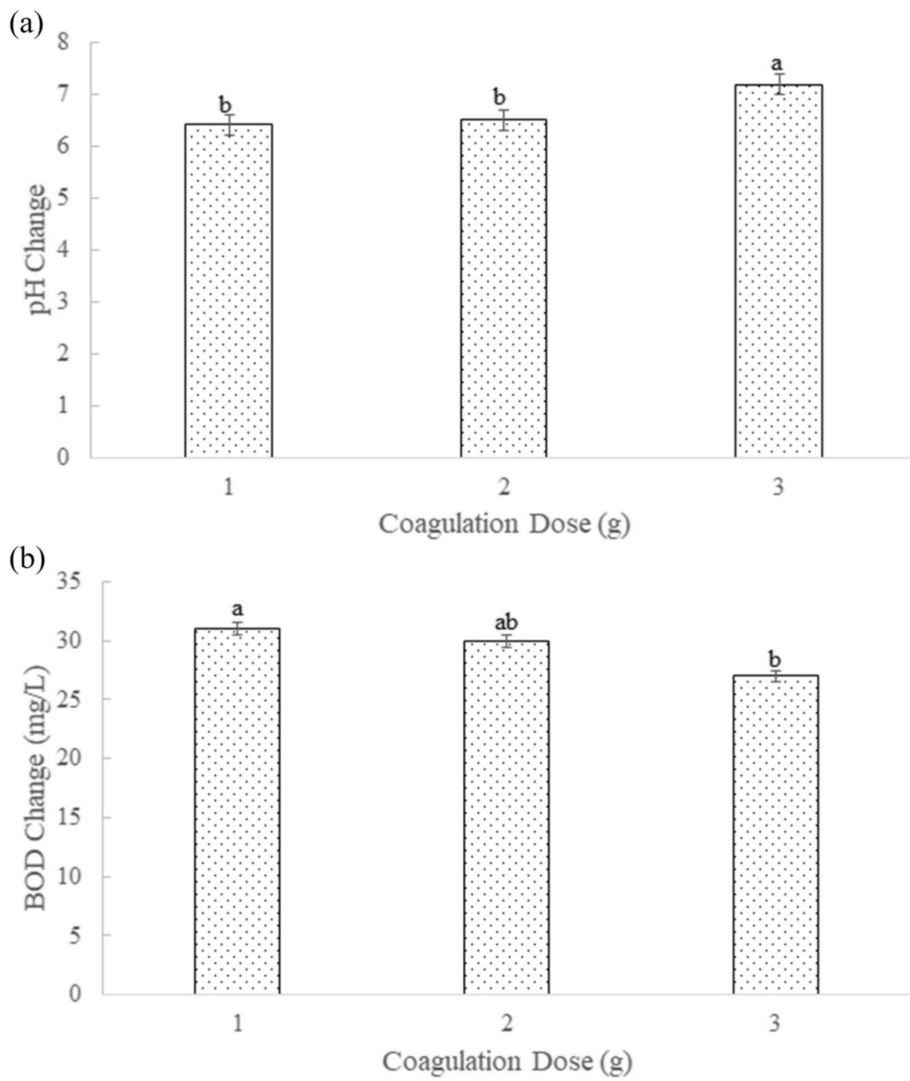

Fig. 1 The (**a**) pH and (**b**) BOD values of the treated wastewater. Means followed by different letters within the same graph are significantly different at $p \leq 0.05$

when pH 6–7 [22]. These negative charges in *C. papaya* coagulants were unable to create flocs if they had the exact charges with colloids. Thus, colloids and coagulants would deny each other. With more *C. papaya* seeds, coagulants from 1 to 3 g did not alter significantly as colloids and coagulants could not react. Thus, the addition of more coagulants would lead to more contamination of the water. A similar finding by Kristanda et al. [21] also explained that adding more coagulants would only pollute the water because the excess coagulants only caused the restabilization colloids.

In addition, reducing TSS from water was governed by charge neutralization and adsorption [26]. If coagulant content surpassed the optimum dosage, the TSS in water could increase because colloids have been precipitated with an optimum dosage. Thus, the excrescent coagulants could cause high TSS in the water when they did not react with conversely charged colloids. The treated TSS values in the *C. papaya* seeds coagulant have yet to meet the Indonesian domestic wastewater quality standard, slightly

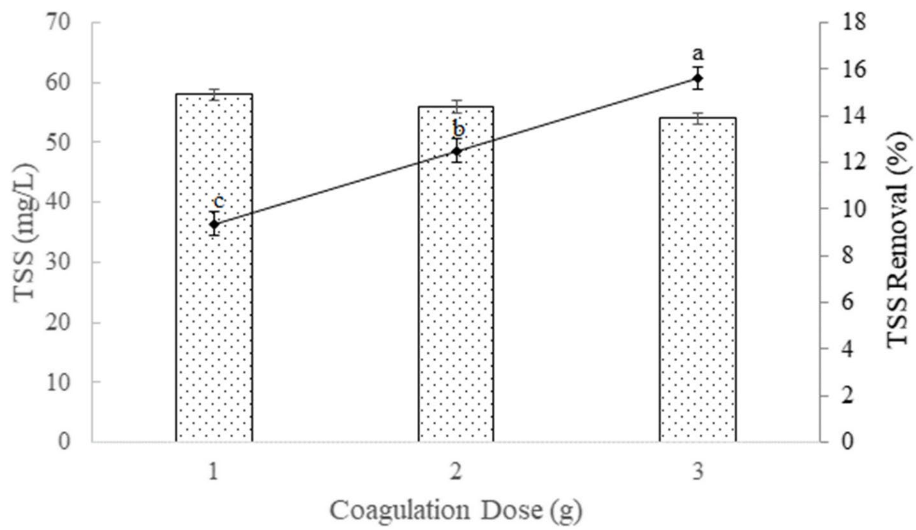

Fig. 2 TSS removal efficiency corresponds to coagulation dose variation

above the permissible limit level of 30 mg/L. In our study, the treated TSS value in wastewater ranged from 54 to 58 mg/L. However, it has been reduced from the initial condition before treatment, which was 64 mg/L. Thus, a higher dosage of coagulant could be considered to obtain the TSS value, which met the Indonesian domestic wastewater quality standard for future studies. The continuous reduction from 1 to 3 g of coagulant dosages experiment showed the potential for a higher reduction of TSS value in wastewater. The result of the TSS removal efficiency ANOVA test in different coagulant dosages showed that statistically significant differences were found among the treatments ($p < 0.05$). The comparison of the before and after treatment data for TSS removal efficiency also showed significant differences ($p < 0.05$) where the values were at optimum dosages in all treatments.

Furthermore, some coagulants only gave a good performance of TSS treatment for low TSS water conditions. The higher the flocculant concentration, the more opposite charge incidents for colloidals occurred. Thus, they tended to repel each other to block the coagulation. As a result of this phenomenon, the efficacy of adsorption relied on the surface morphology, polarity, the adsorbent surface area, pore size distribution, etc. [26]. This notion was in line with a recent study by Kusniawati et al. [23] that explained that *C. papaya* seeds contain natural polymers (e.g., proteins and carbohydrates) that can be used as natural coagulants. Positive and negative charges on proteins can assist the deposition of pollutant particles in water because proteins can initiate the attraction between charges or charge neutralization.

Effect of bio coagulant dosage on COD removal

Before treatment, the initial COD level (145 mg/L) was higher than the Indonesian domestic wastewater quality standard (100 mg/L). After treatment, the COD level was reduced to 128 mg/L using 1 g of coagulant dosage. Then, it showed a significant reduction when we used a 3 g dosage with a COD level decreased to 88 mg/L (Fig. 3). The COD removal after treatment increased when the coagulant dosage increased.

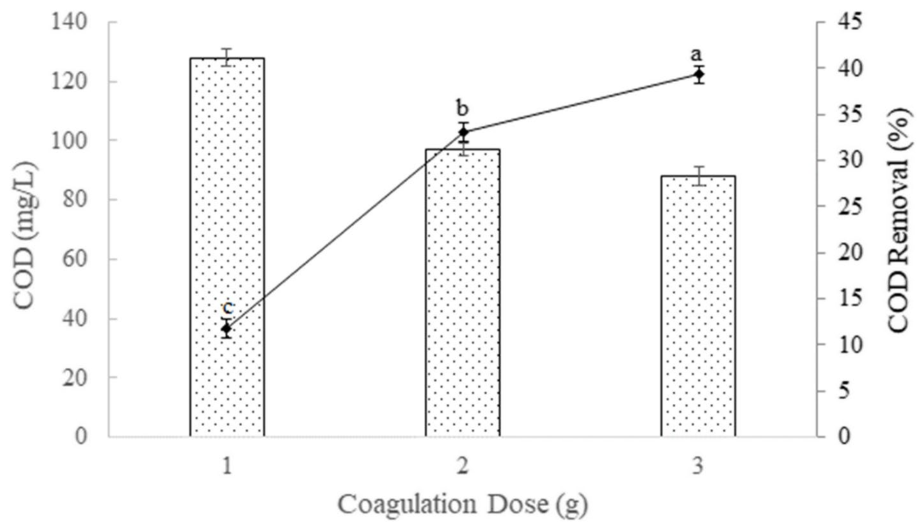

Fig. 3 COD removal efficiency corresponds to coagulation dose variation

The highest COD removal value was observed at 3 g of coagulant treatment, at around 39.3%. Meanwhile, adding 1 g coagulant into wastewater only obtained a COD removal percentage of 11.7% (Fig. 3). However, our study reported a gradual increase in COD removal efficiency after treatment. This result was also found by Handayani et al. [16] in the same study. That study concluded that using *C. papaya* as a coagulant has increased COD removal. The COD is proportional to the total mass of coagulant used. According to our results, the optimum dosage of coagulant for COD removal was 3 g (Fig. 3). The statistical analysis *t*-test exhibited that the before and after treatment data for COD removal in all treatments were significant ($p < 0.05$).

Furthermore, the ANOVA test showed that the mean of COD removal values was statistically significant differences ($p < 0.05$) among the treatments. The reduction of COD values in wastewater samples might be affected by some mechanism that involves the characteristic of *C. papaya* as the coagulant. Firstly, COD removal had a prominent role in decreasing TSS in the wastewater. The adsorption process is an essential mechanism for the process of coagulation. The coagulant from *C. papaya* had several uses of the inter-particle bridge of coagulation and adsorption, for instance, in the adsorption of organic matter. Another possible reduction of COD value was the presence of pollutants in wastewater dominated by charge neutralization and adsorption.

Effect of bio coagulant dosage on phosphate concentration

The pH value is also a prominent factor for the efficiency removal of phosphate using coagulants because the solubility of their precipitate differs with pH. The optimum pH for phosphate removal using the *C. papaya* seeds varied of 5.5–6.5, but in particular wastewater varying from 6 to 9. The predominant phosphate species in the solution are mostly under acidic condition ($H_2PO_4^-$ with pH 2–7) with the relative concentration of 62.5%. Hosni et al. [18] reported that ions in most of coagulants were soluble,thus, they lead to the efficiency of phosphate removal reduced. In this study, after raising the dosage of the *C. papaya* seeds, the pH value is more substantial as compared to the initial pH before treatment. The effect of pH variation on the concentration of phosphate in the water samples could be observed by analyzing the orthophosphate compounds changes. After *C. papaya* seeds were added into the solution, the pH value slightly reduced due to the part of the coagulant precipitated into the hydroxide and hydrogen ions. But when the pH value decreased to less than 5.5, most of ions in the *C. papaya* seeds became soluble. Also, the formation of insoluble phosphate compounds would not occur when it was soluble below pH 6 or above 8 [4]. Contrarily, as the pH value was above 8 after the addition of the *C. papaya* seeds, most of ions became soluble; thus, the efficacy of the coagulation reduced. Additionally, the *C. papaya* seeds contained cationic proteins (positive ions). The bond between positive ions in *C. papaya* seeds and negative ions in the wastewaster samples caused colloid particles and the formation of flocs which could precipitate through gravitational force. Therefore, cations, anions, organic, and inorganic matters also have an important contribution on the exchange for phosphate in the solution.

Figure 4 exhibits the phosphate concentration in the treated wastewater samples. In our study, the phosphate removal was linear to adding coagulant dosage. The highest phosphate removal efficiency was recorded at 3 g of coagulant dosage with 68.4%. The decrease in phosphate removal efficiency after optimum dosage application might be due to the restabilization of colloidal suspensions. There was also the possibility of phosphate removal because aluminum formed aluminum hydroxide flocs that helped in the aluminum phosphate floc sedimentation and other organic matters in the laundry wastewater [4]. Based on the Environmental Protection Organization (EPO) standard for wastewater, the critical limit of phosphate concentration in wastewater was 6 mg/L. Meanwhile, the initial condition of our wastewater samples was 19 mg/L, which was much higher than the EPO standard. Then, after treatment, our study found a significant reduction in phosphate concentration from 19 to 6 mg/L. The ANOVA test also showed that the mean phosphate removal values were statistically significant differences ($p < 0.05$) among the treatments. In addition, the statistical test also obtained that the

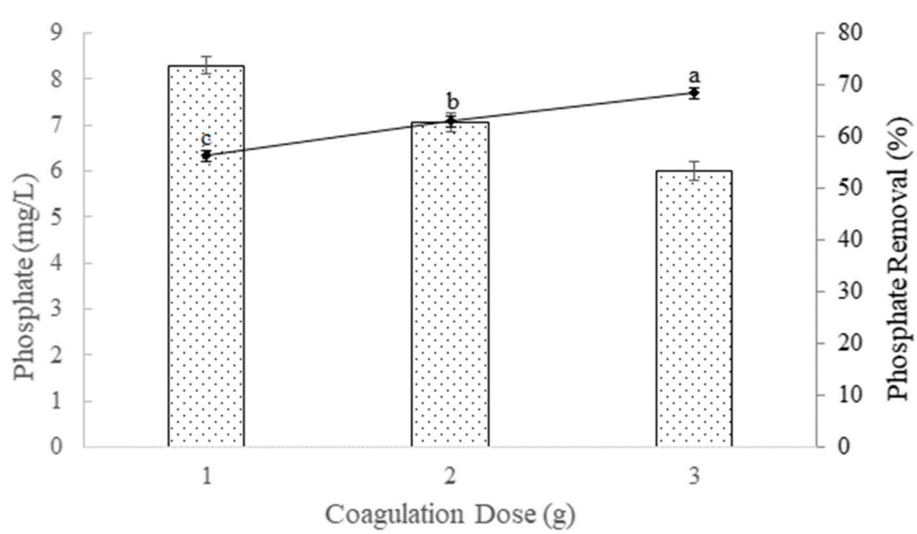

Fig. 4 Phosphate removal efficiency corresponds to coagulation dose variation

Table 2 Laundry wastewater analysis parameters at before and after treatments

Parameter	This study		Previous study [11]		Quality standards
	BT	AT	BT	AT	
pH	7.63	7.19	8.1	6.5	6–9[a]
BOD (mg/L)	33	27	-	-	30[a]
COD (mg/L)	145	88	1160	534	100[a]
TSS (mg/L)	64	54	117.5	78.75	30[a]
Phosphate (mg/L)	19	6	14.29	28.55	6[b]

BT Before treatment, *AT* After treatment

[a] Indonesian domestic wastewater quality standard

[b] Environmental Protection Organization standard

Table 3 Comparison of laundry wastewater treatment using the *C. papaya* seeds and alum products

Parameter	Using *C. papaya* seeds (This study)		Using Alum [34]		Quality standards
	BT	AT	BT	AT	
COD (mg/L)	145	88	1193.06	293.93	100[a]
TSS (mg/L)	64	54	350	34	30[a]
Phosphate (mg/L)	19	6	3.24	0.16	6[b]

BT Before treatment, *AT* After treatment

[a] Indonesian domestic wastewater quality standard

[b] Environmental Protection Organization standard

before and after treatment data for phosphate removal in all treatments were significant ($p < 0.05$). As a whole, the proposed coagulant from *C. papaya* seeds has proven to have some benefits on the wastewater treatment. Table 2 shows the comparison of the results obtained from this study and the previous study using the same coagulant and the wastewater. Most of the water parameters showed a notable reduction in both studies, except for phosphate, where the previous study showed an increase after the addition of coagulant. It might be due to the particle size of *C. papaya* used during the coagulant preparation stage where for our study used 40 mesh while the previous study used 100 mesh. Amin et al. [5] found that the smaller the particle size of coagulant, the better the level of reduction in phosphate concentration. Other studies also assumed the same notion [7].

Furthermore, the *C. papaya* coagulant obtained high percentage removals of phosphate and impurities as compared to alum (Table 3). Utilizing the *C. papaya* seeds to remediate laundry wastewater resulted positive outputs due to environmental friendly and economic of the product used. Therefore, it produces a sustainable, effective, and potent wastewater treatment material. Finally, it can support the small textile industries around Palembang city and other rural areas to treat their own wastewater. However, the use of excess dosage of *C. papaya* seeds coagulant can increase the residual turbidity in the water because the polymer string of *C. papaya* seeds are dominantly related to each other due to the overloaded effect and surface saturation. Then, it will give a negative impact on aquatic environment if the disposal of residual materials is carelessly done [31].

Table 4 FTIR spectrum of *C. papaya* seed coagulant before and after treatments

Wavelength range (cm^{-1})	Before treatment	After treatment	Difference	Remarks
3500–3001	3424.6	3429.9	− 5.3	O–H
	3006.2	-	-	N–H
3000–2001	2923.7	-	-	N–H
	2852.8	-	-	N–H
	2362.7	2362.8	− 0.10	N–H
2000–1501	1744.9	-	-	C = C
	1654.3	-	-	C = C
1500–1001	1463.5	-	-	$=CH_3$
	1161.7	-	-	C-O
	1118.1	1117.7	0.4	C-O
1000–500	721.3	722.4	− 1.1	N–H
	618.6	618.7	− 0.1	N–H

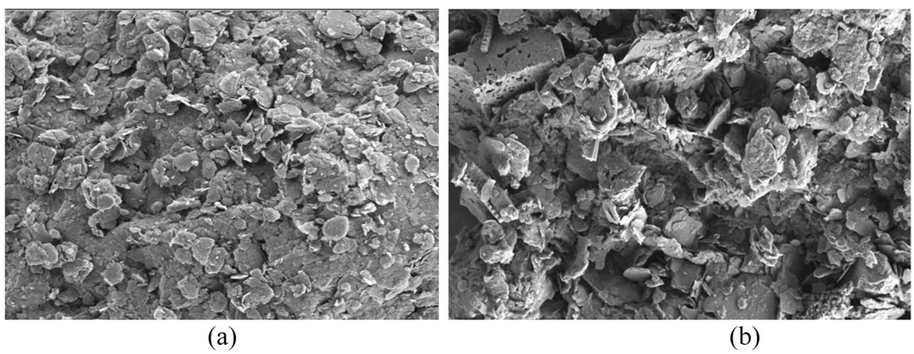

Fig. 5 Surface morphology of *C. papaya* seeds coagulant using SEM before (**a**) and after (**b**) treatments (with magnification × 500)

FTIR and SEM analyses

The peak wavelengths and the corresponding functional groups before and after treatments are depicted in Table 4. Based on Table 4, there was the presence of O–H, C–O, N–H, and $=CH_3$. But after the coagulation process, several of the peaks for O–H, C–O, N–H, and $=CH_3$ functional groups were absent and several peaks were moved. Furthermore, the movement in some peaks might be due to the deformation of the C structures of hemicelluloses, lignin, cellulose, and pectin of the *C. papaya* seeds. According to Zheng et al. [36], the O–H group had a substantial functional part in the surface complex formation with anions, e.g., phosphate. The SEM images of the *C. papaya* seed coagulant before and after treatment are exhibited in Fig. 5. Before the coagulation, the SEM image indicated rough, many gaps, round sphere structures of the coagulant (Fig. 5a). In the case of after coagulation, the circular spheres flocculated and the gaps in the surface have reduced as shown in Fig. 5b. The alteration in the shapes showed the coagulation of pollutants from the wastewater.

Conclusions

This study analyzed *C. papaya* seeds as a bio coagulant for the laundry wastewater treatment. Using *C. papaya* seeds coagulant could be a solution in treating the wastewaters containing high phosphate concentration. Furthermore, the coagulant is eco-friendly, renewable resources, energy efficient, and suitable for the current trends in the wastewater treatment techniques. Overall, the results obtained concluded that the use of the *C. papaya* seeds coagulant showed comparable results in TSS, COD, and phosphate removals with chemical coagulant like alum ($Al_2(SO_4)_3.18H_2O$). Thus, the *C. papaya* seeds can be used as an alternative coagulant product to treat wastewaters. For future works, we suggest to conduct the optimization study regarding the optimum dosage of *C. papaya* seed coagulant. Extending the dosage range (i.e., 1–10 g) will give a new notion which the best dosage of *C. papaya* seeds to treat the wastewaters and also the trend of water quality parameters corresponding to the increased amount of the coagulant.

Abbreviations

ANOVA	Analysis of variance
APHA	American Public Health Association
BOD	Biochemical oxygen demand
C. papaya	*Carica papaya*
COD	Chemical oxygen demand
FTIR	Fourier transform infrared spectroscopy
TSS	Total suspended solids
EPO	Environmental protection organization
SEM	Scanning electron microscope
SMEWW	Standard Methods for the Examination of Water and Wastewater

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Authors' contributions
All authors contributed to the study conception and design. Material preparation, data collection, and analysis were performed by SANV and ZTB. The first draft of the manuscript was written by MR. TEA, SN, LH, and DA contributed in supplying data and analysis. All authors read and approved the fnal manuscript.

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Availability of data and materials
Data will be made available on request.

Declarations

Competing interests
The authors declare no competing interests.

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