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Research Article

The Utilization of Pectin as Natural Coagulant-Aid in Congo Red Dye Removal

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Abstract
Coagulation using inorganic compounds such as aluminum sulfate is commonly used in water-wastewater treatment. However, there are some drawbacks to its utilization, such as a significant decrease in the treated water's pH, non-biodegradable sludge, and a potential negative impact on human mental health (dementia and Alzheimer's). The use of inorganic coagulants can be minimized with the addition of natural-based coagulant-aid such as pectin. In this study, Congo red solution, a model dye substance, was coagulated by varying the pH (3–7) using alum coagulant to determine the best pH for coagulation. At the best pH, pectin was introduced at various doses (0–30 mg/L), and subsequently various dye concentrations (50–100 mg/L). The effect of pectin as coagulant-aid was compared with aluminum sulfate and pectin only; with a response of %removal and sludge volume. It was found that the Congo red dye coagulation had the best %removal at pH 6 indicating a charge neutralization mechanism. The addition of 15 mg/L pectin at an aluminum sulfate dose of 30 mg/L resulted in 97.7% dye removal with a sludge volume of 14 mL/L at a Congo red concentration of 50 mg/L. This value is higher compared to those of aluminum sulfate and pectin only which gave 75.6 and 3.19% removals, respectively. Furthermore, the addition of pectin as a natural coagulant-aid could halve the sludge volume due to the formation of denser flocs. The results show a promising potential of pectin as a natural coagulant-aid in water-wastewater treatment.

Keywords: Congo red, coagulant-aid, natural coagulant, pectin

1. INTRODUCTION

Coagulation and flocculation are the most commonly used methods for water and wastewater treatment due to their high efficiency and low cost [1]. Furthermore, inorganic coagulants such as alum ($Al_2(SO_4)_3$), $FeCl_3$, and PAC (poly(aluminum chloride)) are widely used due to their ability to reduce color intensity, organic content and turbidity [2]. Aside from its high efficiency, the use of inorganic coagulants has several limitations, including lowering water alkalinity, producing large amounts of sludge, and the possibility of adverse effects on both human health and the environment [3]. As a result, natural-based coagulants have been explored as an alternative to reduce or substitute the use of inorganic coagulants.

Natural coagulants could be classified as protein, polysaccharide, or polyphenol, based on their active coagulating agent [4]. Protein-based coagulant that comes from beans and legume extract has been extensively studied. However, the extraction and purification steps turned out to form a hindrance to its wide application and commercialization [3]. On the other hand, polyphenol-based coagulant has also been investigated. Even so, the amount of research is relatively limited, thus more investigation in the future is needed [5]. Lastly, polysaccharide is known as the most abundant polymer found in nature [6], making it an interesting source to be employed as natural coagulant.

Previous studies have utilized various polysaccharide sources as natural coagulants, such as starch, gum, chitosan, and the like. Starch from rice, sago, and corn has been explored as natural coagulant to treat textile wastewater, landfill leachate, and agro-industrial wastewater [7]-[10]. This starch gave a good coagulation performance. However, the utilization of starch from food sources poses a potential conflict of interest, thus limiting its commercial application [11]. Gum from various sources also has been utilized as natural coagulant and coagulant-aid [12]-[15]. Fruit industrial waste is an alternative non-food source that can be considered for use in the application as well. This fruit waste which is commonly generated as agricultural byproduct has a high potential for various applications due to its various compounds [16]. This waste contains active ingredients in the

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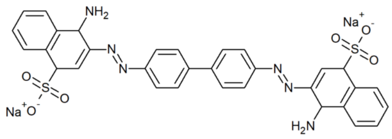
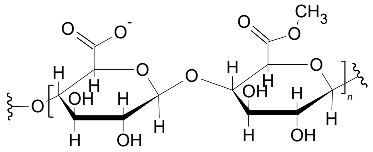
Table 1. General properties of Congo red and pectin

Properties	Congo red	Pectin
Molecular formula	$C_{32}H_{22}N_6Na_2O_6S_2$	$C_6H_{10}O_7$ (monomer)
Molecular weight (g/mol)	696.7 g/mol	194.1 (monomer)
Molecular structure		

form of polysaccharides such as pectin and starch, which have shown their potential as coagulant and coagulant-aid [17][18].

Compared to starch, pectin is a good alternative as coagulant-aid because pectin can be used as it is, without any modification needed. Pectin has also been commercialized and widely used for various applications such as food additives [19], medical applications [20], and nutraceuticals [21]. Pectin is a heteropolysaccharide structure that is composed of α-D-(1,4) galacturonan and rhamnogalacturonan with branches consisting of neutral sugar such as rhamnose, arabinose, galactose, and xylose [22]. Moreover, pectin can be differentiated as high and low-methoxy pectin, based on the degree of esterification. High-methoxy pectin with a degree of esterification of over 50% can be made to interact with sugar to form gels, and is commonly used in jam-making [23]. On the other hand, low-methoxy pectin has a degree of esterification lower than 50%. It can interact with multivalent cations via the egg box model [23], making it suitable for application as coagulant-aid.

Furthermore, compared to synthetic polymers, the utilization of pectin could become a good alternative due to its highly biodegradable and non-toxic properties. For example, polyacrylamide and acrylamide copolymers (anionic polymers) are commonly used as flocculant with a high molecular weight, being very stable, readily soluble in water, and very effective at a low dosage. However, this polymer is hardly biodegradable with a concern of the polymer and monomer's toxicity [24]. Another example is poly(diallydimethylammonium chloride) (polyDADMAC), a cationic polymer, was proven to be chronically toxic to *Ceriodaphnia dubia* [25], emphasizing the toxicity of synthetic polymer to aquatic organisms and the ecosystem.

Previously, pectin had been used as a coagulant or coagulant-aid to remove heavy metals [26] and turbidity [27][28] in synthetic wastewater. However, to the best of the authors' knowledge, the study of pectin as coagulant-aid to treat synthetic dye wastewater has never been investigated. In this study, the application of low-methoxy pectin as a coagulant-aid and alum has been used to treat synthetic Congo red as a model wastewater. Several variables that influence the coagulation, such as pH, coagulant-aid dosage, and dye concentration have also been examined.

2. MATERIALS AND METHODS

2.1. Materials

Congo red dye was obtained from Sigma-Aldrich, while alum (technical grade) and low-methoxy pectin (food grade) was purchased from a local shop in Bandung, West Java, Indonesia. All chemicals were used as obtained, without any further treatment. The general properties of Congo red and pectin are presented in Table 1.

2.2. Jar Test Experiment

The coagulation study of Congo red was conducted by using a jar test apparatus. A stock Congo red solution (1 g/L) was prepared and subsequently diluted using distilled water to obtain the desired Congo red concentration. The variations of coagulation study are presented in Table 2. The initial pH of the solution was adjusted by using 0.1 M HCl or NaOH and measured by using a calibrated pH meter (Lutron ph-208). The coagulation was accomplished by mixing Congo red, alum and followed by pectin solutions at rapid mixing (200 rpm, 2 min), followed by slow mixing (40 rpm, 20 min), and settling for 1 h. The initial

Table 2. Variations applied in this study

Variations	pH	Alum dose (mg/L)	Pectin dose (mg/L)	Congo red concentration (mg/L)
pH study	3, 4, 5, 6, 7	50	0	50
Coagulant-aid study	Best pH	30	0, 5, 10, 15, 20, 25, 30	50
Initial Congo red concentration	Best pH	30	Best pectin dose	50, 60, 70, 80, 90, 100
		30	0	
		0	Best pectin dose	

(C_i; mg/L) and final (C_f; mg/L) concentrations of Congo red were measured using a visible spectrophotometer (Thermoscientific Genesys 30) at its maximum wavelength (510 nm). The removal percentage (%removal) was calculated using Equation 1. Furthermore, the sludge volume (mL/L) was measured using an Imhoff cone after 1 h of settling and calculated using Equation 2. All experiments were carried out in duplicate and standard deviations as provided in each figure, where applicable.

$$\%\,removal = \frac{(C_i - C_f)}{C_f} \times 100\% \quad (1)$$

$$sludge\ volume\left(\frac{mL}{L}\right) = \frac{V\ sludge\ (mL)}{V\ wastewater\ (L)} \quad (2)$$

3. RESULTS AND DISCUSSIONS

3.1. The Effect of pH Levels on Coagulation

The profile of Congo red dye concentration and sludge volume in coagulation was observed at different pH variations (3, 4, 5, 6, and 7) with the coagulant dose and Congo red dye concentration fixed at 50 mg/L. The obtained results are shown in Figure 1. It can be observed in Figure 1 that with the increase of pH, the Congo red removal also increased until pH 6, before decreasing at pH 7. The highest %removal was obtained at the initial pH of 6 with 92% and a sludge volume of 20 mL/L. In the pH range of 3–6, the Congo red would be positively charged, as the Congo red zero charge is around pH 2. Furthermore, around pH 5 and 6, most of the soluble alum species were in the form of Al^{3+}, $AlOH^{2+}$ and $Al(OH)_2^+$ [29] which could neutralize the negatively charged Congo red molecules. With the dye molecule's neutral charge, the electrostatic repulsion decreases, allowing the formation of flocs. A further increase in pH resulted in hydrolyzation of the alum, forming $Al(OH)_3$ and $Al(OH)_4^-$ while decreasing the positively charged alum species, thus preventing charge neutralization to occur [30]. This reduces the dye's coagulation efficiency at pH 7, resulting in a decrease in sludge volume as well. The result obtained in this study is

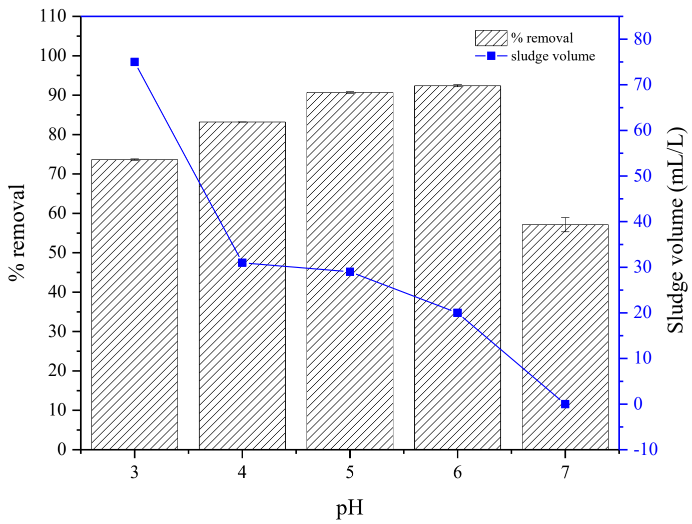


Figure 1. Effect of pH on %removal and sludge volume

similar to previous research studies that reported that the best performance of alum coagulation was achieved at pH 6 [31]-[33].

3.2. The Effect of Coagulant-aid Dose on Coagulation

The effect of the coagulant-aid dose on the % removal of Congo red is presented in Figure 2. The dose of coagulant-aid was varied with the dye concentration fixed at 50 mg/L and the alum dose at 30 mg/L. The amount of alum used in this variation was decreased to 30 mg/L in order to observe the significance of the role pectin plays as a coagulant-aid. It may be observed that an increase in Congo red removal occurred with the addition of pectin until 15 mg/L which gave a 97.0% removal; compared to alum which only gave a 76.0% removal. Further addition of pectin did not increase the %removal of Congo red.

This result clearly indicates that pectin as a coagulant-aid works synergistically with alum in supporting the floc formation. This is closely related to the complex formation that occurs between pectin with small flocs formed previously from the interaction of Congo red and alum. Coagulation that occurs between the alum coagulant and Congo with a charge neutralization mechanism will produce small flocs [34]. Pectin which is subsequently added as a coagulant-aid could act as a particle bridge that helps the formation of larger flocs, resulting in higher coagulation performance. The amount of pectin that was gradually raised until it was over the optimal level did not give a significant increase in the %removal, instead a slight decrease of %removal and increase sludge volume was observed. This is possible due to the over-addition of pectin that could lead to an increase of zeta potential, making a smaller floc formation [35]. In turn, this could lead to more porous and voluminous sludge. Similar observations have been reported by previous researchers [15][36][37].

3.3. The Effect of Dye Initial Concentration on Coagulation

The profile of the %removal of Congo red dyes was observed in various initial concentrations of Congo red dyes (50–100 mg/L), with a fixed dose of alum (30 mg/L) and pectin (15 mg/L) at the best pH of 6. The result is presented in Figure 3.

At a dosage of 15 mg/L, pectin had a very poor coagulant activity and was unable to coagulate the dye molecules. This is apparent from the %removal, which amounted to a mere 3.19% and fell as the initial dye concentration rose. Pectin typically has a negative high zeta potential value at pH 6, which is around -25 mV [38]. In addition, Congo red has an isoelectric point around pH 2 [39] that would be negatively charged as well at pH 6. Based on these facts, it can be concluded that pectin does not act as the active coagulating agent in this study.

Furthermore, it can be observed that with the increase of Congo red concentration, the %removal and sludge formation were also decreasing. This is possible due to the inadequacy of active coagulating sites to neutralize the dye molecules with the increase of Congo red concentration, as the interaction between coagulant and colloid has been

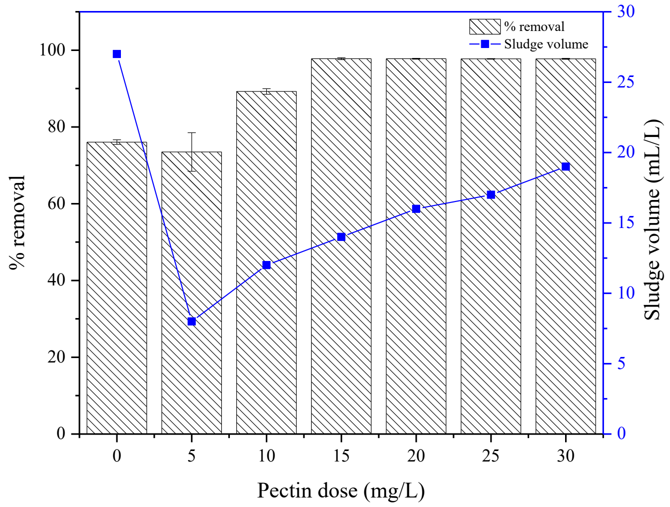


Figure 2. Effect of pectin dose on %removal and sludge volume

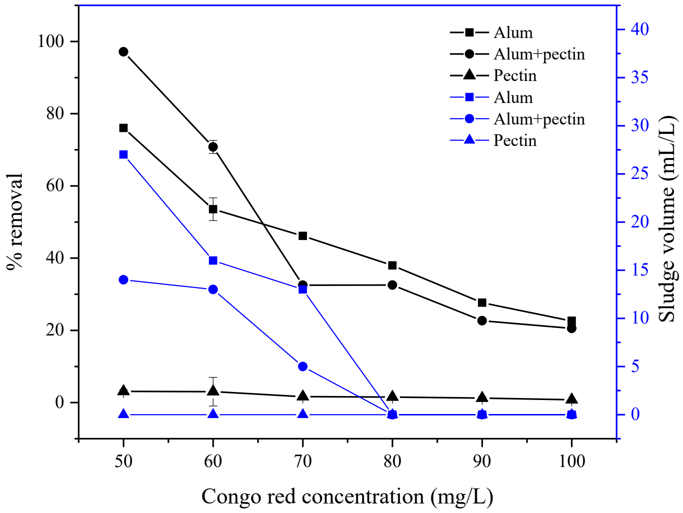


Figure 3. Effect of initial dye concentration on %removal and sludge volume

reported as stoichiometric interaction. The effect of pectin as coagulant-aid is discernible at the initial Congo red concentration of 50–60 mg/L that gave %removal of 97.1 and 70.8% respectively. This value increased significantly when compared to the one measured without using pectin which gave 76.0 and 53.5% removals at the same initial Congo red concentration. This increase confirmed a synergistic effect between alum and pectin that resulted in bigger observed flocs (Figure 4) and higher Congo red removal. The possible interaction between Congo red, alum, and pectin is illustrated in Figure 5. The negatively charged sulfonate groups in the Congo red molecule would be neutralized by the positive Al^{3+}, while the carboxylate groups in the pectin structure could also interact with Al^{3+}, resulting in a particle bridging effect (Figure 5.a). This bridging mechanism could explain the formation of bigger flocs, as observed in Figure 5.b, which resulted in a lower sludge volume and higher removal. Similar results of polysaccharides that act as a bridge between smaller flocs have been reported previously [31][34][40].

The results obtained in this study are compared with other coagulants for Congo red removal, presented in Table 3. It can be observed that the coagulation performance of alum with the aid of pectin could give a comparable coagulation performance with other coagulants reported in the background literature. Furthermore, it can be seen that a smaller alum dosage (30 mg/L) is needed to obtain a 97% removal with the 15 mg/L pectin addition, compared to research conducted by [41] where 100 mg/L alum was required.

4. CONCLUSIONS

In this study, low-methoxy pectin has been

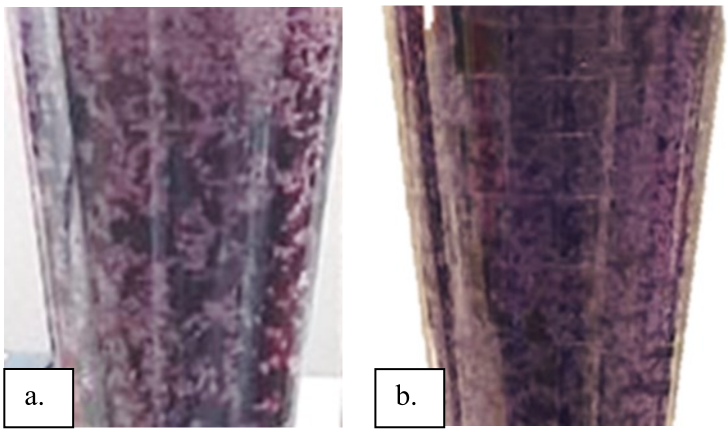


Figure 4. Observed flocs formation with 15 mg/L pectin (a) and without pectin (b) at pH 6. [alum] = 30 mg/L. [Congo red] = 50 mg/L

Table 3. Comparison of various coagulants for Congo red removal

Coagulant		Coagulant-aid		pH	Congo red concentration (mg/L)	% removal	Reference
Type	Dosage (mg/L)	Type	Dosage (mg/L)				
$Al_2(SO_4)_3$	30	Pectin	15	6	50	97.1	This study
$Al_2(SO_4)_3$	30	-	-	6	50	76.0	This study
$CaCl_2$	4,000	Alginate	20	4	50	92.2	[42]
$FeCl_3$	160	Galactomannan	80	6	20	90.0	[15]
$FeCl_3$	10	*A. tetragonus*	300	3	500	92.0	[30]
Polyaluminium chloride	15	-	-	3	50	81.2	[40]
Chitosan	25	-	-	7	200	66.0	[43]
$Al_2(SO_4)_3$ nanoparticles	100			7	50	99.5	[41]

successfully utilized as a coagulant-aid in Congo red dye removal. At various initial pHs, it was found that Congo red removal increases from pH 3–6 with pH 6 as the best pH for coagulation via a charge neutralization mechanism. The addition of pectin could assist the Congo red coagulation process. The addition of 15 mg/L of pectin gave a 97% Congo red removal, compared to the 76% removal when only alum was used as the coagulant. Overdosing of pectin could result in colloid re-stabilization that makes a smaller floc formation and increases the sludge volume. At various initial Congo red concentrations and fixed coagulant and coagulant-aid doses, the effect of pectin addition could increase the Congo red removal at an initial concentration of 50–60 mg/L, which gave an approximately 20% increase of removal compared to alum only. At a higher Congo red concentration, the removal became lower than the one without pectin. This might happen due to an inadequacy of alum at a fixed coagulant dosage with the increase of Congo red concentration. This condition could lead to a competitive interaction of alum-Congo red and alum-pectin, where both Congo red and pectin are negatively charged at the coagulation condition. The results of this research study show that low-methoxy pectin is a potential coagulant-aid that could work synergistically with alum in the coagulation process. Further studies are required to observe the potential of the combination between alum and pectin in other types of wastewaters, real wastewater, and optimization of the variables as well.

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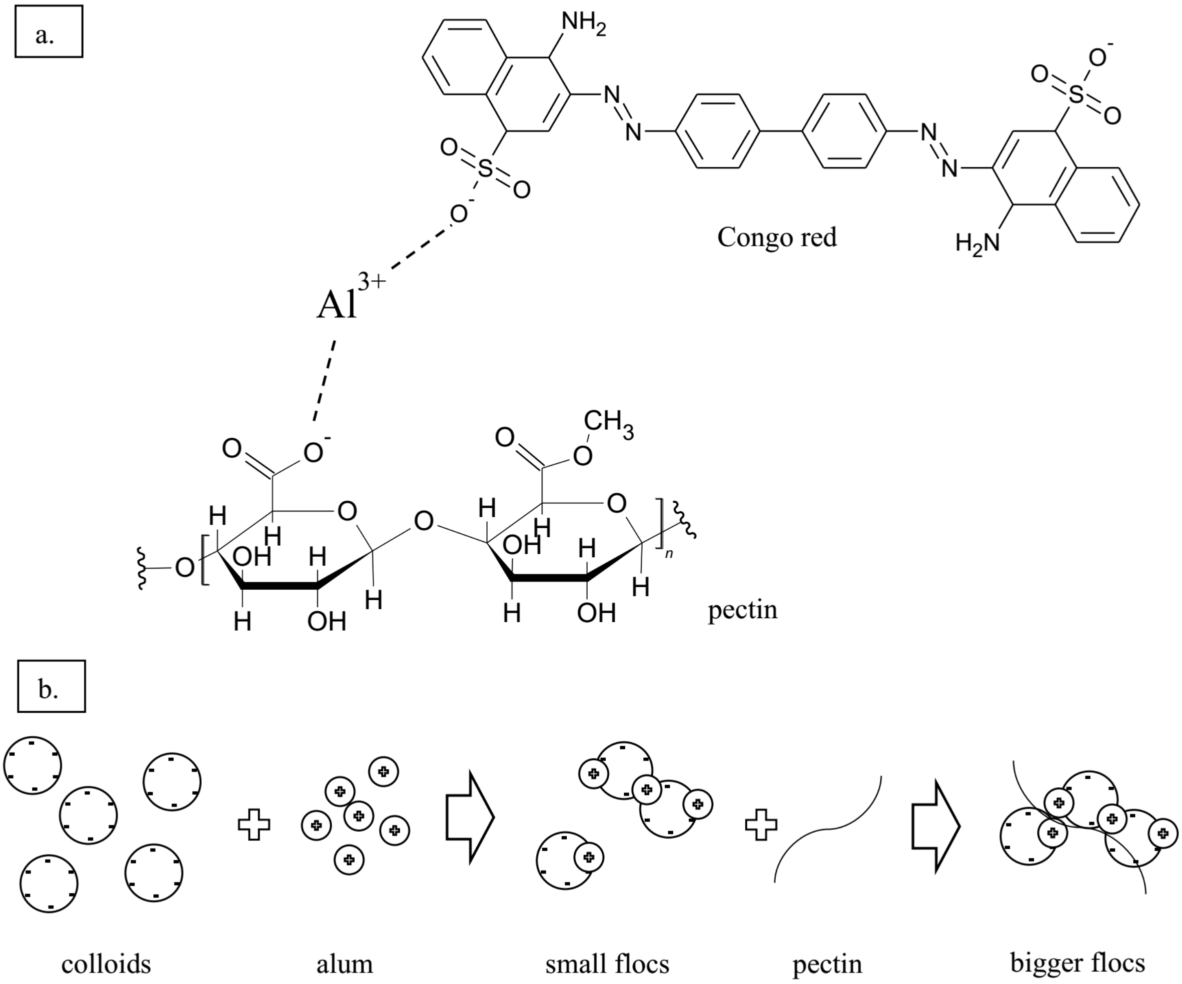

Figure 5. Possible interaction between Al^{3+}–Congo red and Al^{3+}–pectin (a) and the particle bridging of flocs by pectin (b) (Reused with permission from [40]; Copyright © Elsevier license number 5524610093660)

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Conflicts of Interest

The author(s) declare no conflict of interest.

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