

ACCEPTED MANUSCRIPT • **OPEN ACCESS**

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

To cite this article before publication: F. M. K. Haryanto, A. V. M. Rumondor, H. Kristianto, S. Prasetyo, and A. K. Sugih. (2023). *J. Multidiscip. Appl. Nat. Sci.* in press. <https://doi.org/10.47352/jmans.2774-3047.179>.

Manuscript version: Accepted Manuscript

Accepted Manuscript is “the version of the article accepted for publication including all changes made as a result of the peer review process, and which may also include the addition to the article by Pandawa Institute of a header, an article ID, a cover sheet and/or an ‘Accepted Manuscript’ watermark, but excluding any other editing, typesetting or other changes made by Pandawa Institute and/or its licensors”

This Accepted Manuscript is © 2023 **The Author(s). Published by Pandawa Institute**



As the Version of Record of this article is going to be / has been published on a gold open access basis under a CC BY 4.0 International License, this Accepted Manuscript is available for reuse under a CC BY 4.0 International License immediately.

Everyone is permitted to use all or part of the original content in this article, provided that they adhere to all the terms of the license <https://creativecommons.org/licenses/by/4.0/>.

Although reasonable endeavors have been taken to obtain all necessary permissions from third parties to include their copyrighted content within this article, their full citation and copyright line may not be present in this Accepted Manuscript version. Before using any content from this article, please refer to the Version of Record on Pandawa Institute once published for full citation and copyright details, as permissions may be required. All third-party content is fully copyright protected and is not published on a gold open access basis under a CC BY license, unless that is specifically stated in the figure caption in the Version of Record.

View the [article online](#) for updates and enhancements.

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

Felicia M K Haryanto¹⁾; Andranysa V M Rumondor¹⁾; Hans Kristianto^{1,*)};
Susiana Prasetyo¹⁾; Asaf K Sugih¹⁾

¹ *Department of Chemical Engineering, Parahyangan Catholic University, Bandung-40141
(Indonesia)*

*Correspondence: hans.kristianto@unpar.ac.id

ORCIDs:

First AUTHOR : <https://orcid.org/0000-0003-4148-6097>

Second AUTHOR : <https://orcid.org/0000-0002-0017-5623>

Third AUTHOR : <https://orcid.org/0000-0003-4747-4361>

Fourth AUTHOR : <https://orcid.org/0000-0001-5839-0054>

Fifth AUTHOR : <https://orcid.org/0000-0003-0478-540X>

ACKNOWLEDGEMENT

This study is funded by Parahyangan Catholic University's Centre of Research and Community Service with contract No III/LPPM/2023-02/27-P. The authors are thankful for the funding given.

AUTHOR CONTRIBUTIONS

Conceptualization, methodology, investigation, writing – original draft: FMKH, AVMR. Both authors contributed equally to this work. Resources, writing – review & editing, supervision, funding acquisition: HK, SP, and AKS.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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

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 ($\text{Al}_2(\text{SO}_4)_3$), 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

1 both human health and the environment [3]. As a result, natural-based coagulants have been
2 explored as an alternative to reduce or substitute the use of inorganic coagulants.

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

11 Previous studies have utilized various polysaccharide sources as natural coagulants, such
12 as starch, gum, chitosan, and the like. Starch from rice, sago, and corn has been explored as
13 natural coagulant to treat textile wastewater, landfill leachate, and agro-industrial wastewater
14 [7]-[10]. This starch gave a good coagulation performance. However, the utilization of starch
15 from food sources poses a potential conflict of interest, thus limiting its commercial application
16 [11]. Gum from various sources also has been utilized as natural coagulant and coagulant-aid
17 [12]-[15]. Fruit industrial waste is an alternative non-food source that can be considered for
18 use in the application as well. This fruit waste which is commonly generated as agricultural
19 byproduct has a high potential for various applications due to its various compounds [16]. This
20 waste contains active ingredients in the form of polysaccharides such as pectin and starch,
21 which have shown their potential as coagulant and coagulant-aid [17][18].

22 Compared to starch, pectin is a good alternative as coagulant-aid because pectin can be
23 used as it is, without any modification needed. Pectin has also been commercialized and widely
24 used for various applications such as food additives [19], medical applications [20], and
25 nutraceuticals [21]. Pectin is a heteropolysaccharide structure that is composed of α -D-(1,4)
26 galacturonan and rhamnogalacturonan with branches consisting of neutral sugar such as
27 rhamnose, arabinose, galactose, and xylose [22]. Moreover, pectin can be differentiated as high
28 and low-methoxy pectin, based on the degree of esterification. High-methoxy pectin with a
29 degree of esterification of over 50% can be made to interact with sugar to form gels, and is
30 commonly used in jam-making [23]. On the other hand, low-methoxy pectin has a degree of
31 esterification lower than 50%. It can interact with multivalent cations via the egg box model
32 [23], making it suitable for application as coagulant-aid.

33 Furthermore, compared to synthetic polymers, the utilization of pectin could become a
34 good alternative due to its highly biodegradable and non-toxic properties. For example,

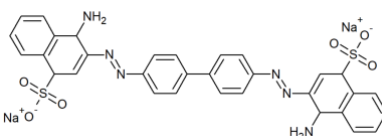
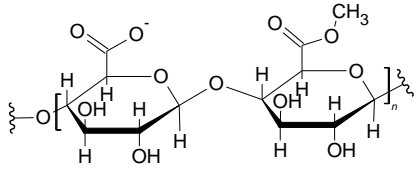
1 polyacrylamide and acrylamide copolymers (anionic polymers) are commonly used as
 2 flocculant with a high molecular weight, being very stable, readily soluble in water, and very
 3 effective at a low dosage. However, this polymer is hardly biodegradable with a concern of the
 4 polymer and monomer's toxicity [24]. Another example is poly(diallyldimethylammonium
 5 chloride) (polyDADMAC), a cationic polymer, was proven to be chronically toxic to
 6 *Ceriodaphnia dubia* [25], emphasizing the toxicity of synthetic polymer to aquatic organisms
 7 and the ecosystem.

8 Previously, pectin had been used as a coagulant or coagulant-aid to remove heavy metals
 9 [26] and turbidity [27][28] in synthetic wastewater. However, to the best of the authors'
 10 knowledge, the study of pectin as coagulant-aid to treat synthetic dye wastewater has never
 11 been investigated. In this study, the application of low-methoxy pectin as a coagulant-aid and
 12 alum has been used to treat synthetic Congo red as a model wastewater. Several variables that
 13 influence the coagulation, such as pH, coagulant-aid dosage, and dye concentration have also
 14 been examined.

16 2. MATERIALS AND METHOD

17
 18 *2.1. Materials.* Congo red dye was obtained from Sigma-Aldrich, while alum (technical
 19 grade) and low-methoxy pectin (food grade) was purchased from a local shop in Bandung,
 20 West Java, Indonesia. All chemicals were used as obtained, without any further treatment. The
 21 general properties of Congo red and pectin are presented in Table 1.

22
 23 **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		

1 2.2. *Jar test experiment.* The coagulation study of Congo red was conducted by using a
 2 jar test apparatus. A stock Congo red solution (1 g/L) was prepared and subsequently diluted
 3 using distilled water to obtain the desired Congo red concentration. The variations of
 4 coagulation study are presented in Table 2. The initial pH of the solution was adjusted by using
 5 0.1 M HCl or NaOH and measured by using a calibrated pH meter (Lutron ph-208). The
 6 coagulation was accomplished by mixing Congo red, alum and followed by pectin solutions at
 7 rapid mixing (200 rpm, 2 min), followed by slow mixing (40 rpm, 20 min), and settling for 1
 8 h. The initial (C_i ; mg/L) and final (C_f ; mg/L) concentrations of Congo red were measured using
 9 a visible spectrophotometer (Thermoscientific Genesys 30) at its maximum wavelength (510
 10 nm). The removal percentage (%removal) was calculated using Equation 1. Furthermore, the
 11 sludge volume (mL/L) was measured using an Imhoff cone after 1 h of settling and calculated
 12 using Equation 2. All experiments were carried out in duplicate and standard deviations as
 13 provided in each figure, where applicable.

$$15 \quad \% \text{ removal} = \frac{(C_i - C_f)}{C_f} \times 100\% \quad (1)$$

$$17 \quad \text{sludge volume} \left(\frac{\text{mL}}{\text{L}} \right) = \frac{V_{\text{sludge}} (\text{mL})}{V_{\text{wastewater}} (\text{L})} \quad (2)$$

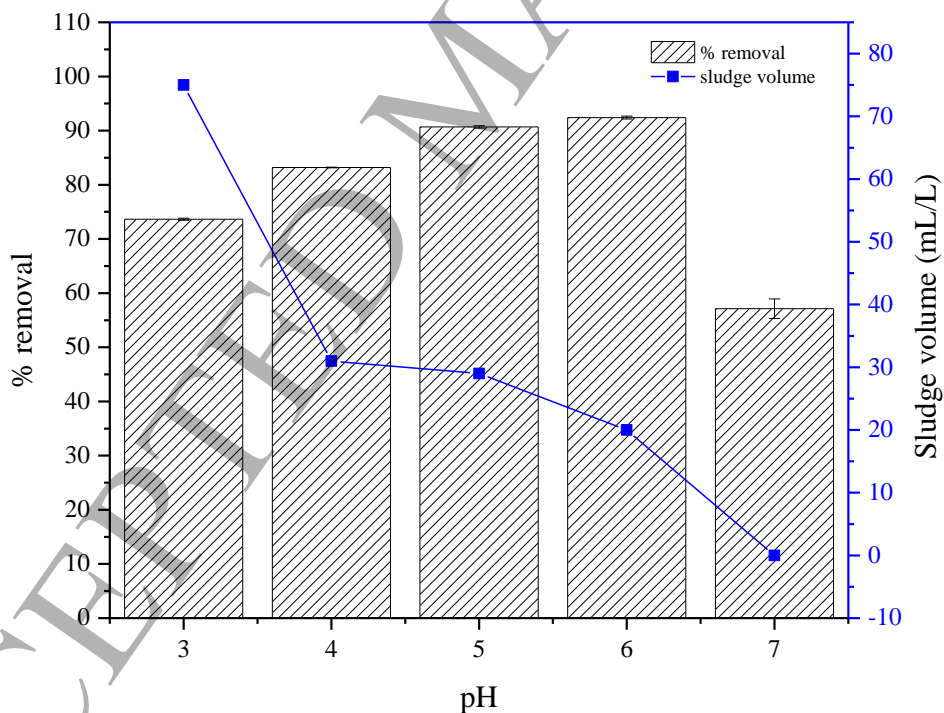
19 **Table 2.** Variations applied in this study

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,
		30	0	100
		0	Best pectin dose	

21 3. RESULTS AND DISCUSSION

23 3.1. *The effect of pH levels on coagulation.* The profile of Congo red dye concentration
 24 and sludge volume in coagulation was observed at different pH variations (3, 4, 5, 6, and 7)

1 with the coagulant dose and Congo red dye concentration fixed at 50 mg/L. The obtained results
 2 are shown in Figure 1 below. It can be observed in Figure 1 that with the increase of pH, the
 3 Congo red removal also increased until pH 6, before decreasing at pH 7. The highest %removal
 4 was obtained at the initial pH of 6 with 92% and a sludge volume of 20 mL/L. In the pH range
 5 of 3–6, the Congo red would be positively charged, as the Congo red zero charge is around pH
 6 2. Furthermore, around pH 5 and 6, most of the soluble alum species were in the form of Al^{3+} ,
 7 $AlOH^{2+}$ and $Al(OH)_2^+$ [29] which could neutralize the negatively charged Congo red
 8 molecules. With the dye molecule's neutral charge, the electrostatic repulsion decreases,
 9 allowing the formation of flocs. A further increase in pH resulted in hydrolyzation of the alum,
 10 forming $Al(OH)_3$ and $Al(OH)_4^-$ while decreasing the positively charged alum species, thus
 11 preventing charge neutralization to occur [30]. This reduces the dye's coagulation efficiency
 12 at pH 7, resulting in a decrease in sludge volume as well. The result obtained in this study is
 13 similar to previous research studies that reported that the best performance of alum coagulation
 14 was achieved at pH 6 [31]-[33].

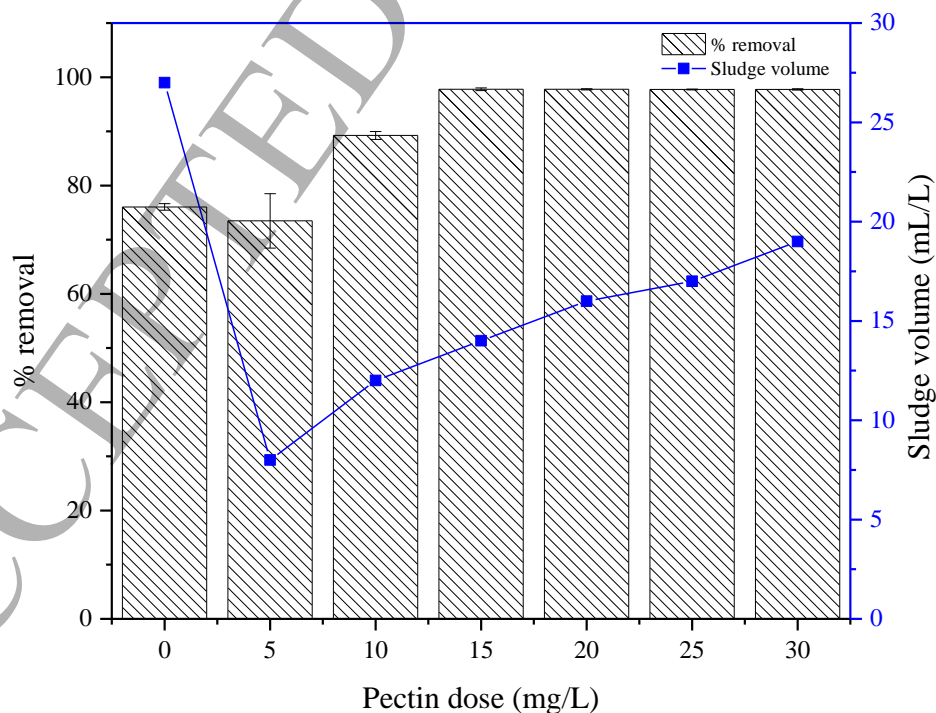


15 **Figure 1.** Effect of pH on %removal and sludge volume
 16

17
 18 **3.2. The effect of coagulant-aid dose on coagulation.** The effect of the coagulant-aid dose
 19 on the %removal of Congo red is presented in Figure 2. The dose of coagulant-aid was varied

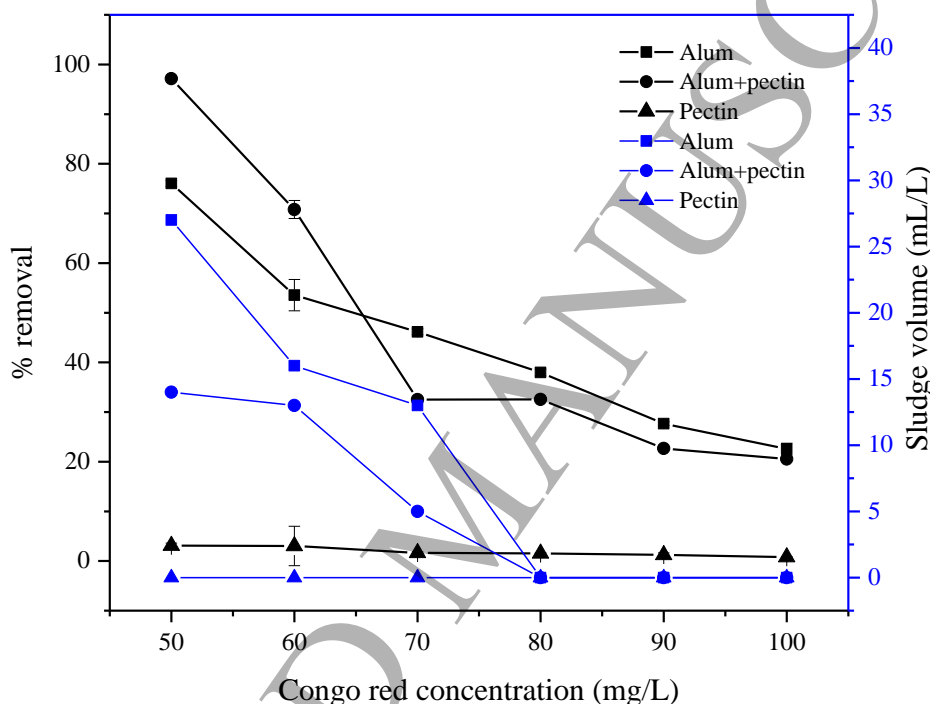
1 with the dye concentration fixed at 50 mg/L and the alum dose at 30 mg/L. The amount of alum
2 used in this variation was decreased to 30 mg/L in order to observe the significance of the role
3 pectin plays as a coagulant-aid. It may be observed that an increase in Congo red removal
4 occurred with the addition of pectin until 15 mg/L which gave a 97.0% removal; compared to
5 alum which only gave a 76.0% removal. Further addition of pectin did not increase the
6 %removal of Congo red.

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



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

2
3 3.3. *The effect of dye initial concentration on coagulation.* The profile of the %removal
4 of Congo red dyes was observed in various initial concentrations of Congo red dyes (50–100
5 mg/L), with a fixed dose of alum (30 mg/L) and pectin (15 mg/L) at the best pH of 6. The result
6 is presented in Figure 3.

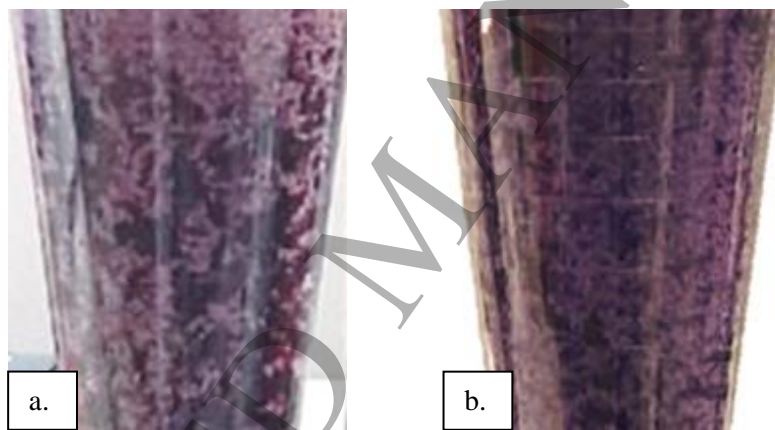


7
8 **Figure 3** Effect of initial dye concentration on %removal and sludge volume

9
10 At a dosage of 15 mg/L, pectin had a very poor coagulant activity and was unable to
11 coagulate the dye molecules. This is apparent from the %removal, which amounted to a mere
12 3.19% and fell as the initial dye concentration rose. Pectin typically has a negative high zeta
13 potential value at pH 6, which is around -25 mV [38]. In addition, Congo red has an isoelectric
14 point around pH 2 [39] that would be negatively charged as well at pH 6. Based on these facts,
15 it can be concluded that pectin does not act as the active coagulating agent in this study.

16 Furthermore, it can be observed that with the increase of Congo red concentration, the
17 %removal and sludge formation were also decreasing. This is possible due to the inadequacy
18 of active coagulating sites to neutralize the dye molecules with the increase of Congo red
19 concentration, as the interaction between coagulant and colloid has been reported as

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



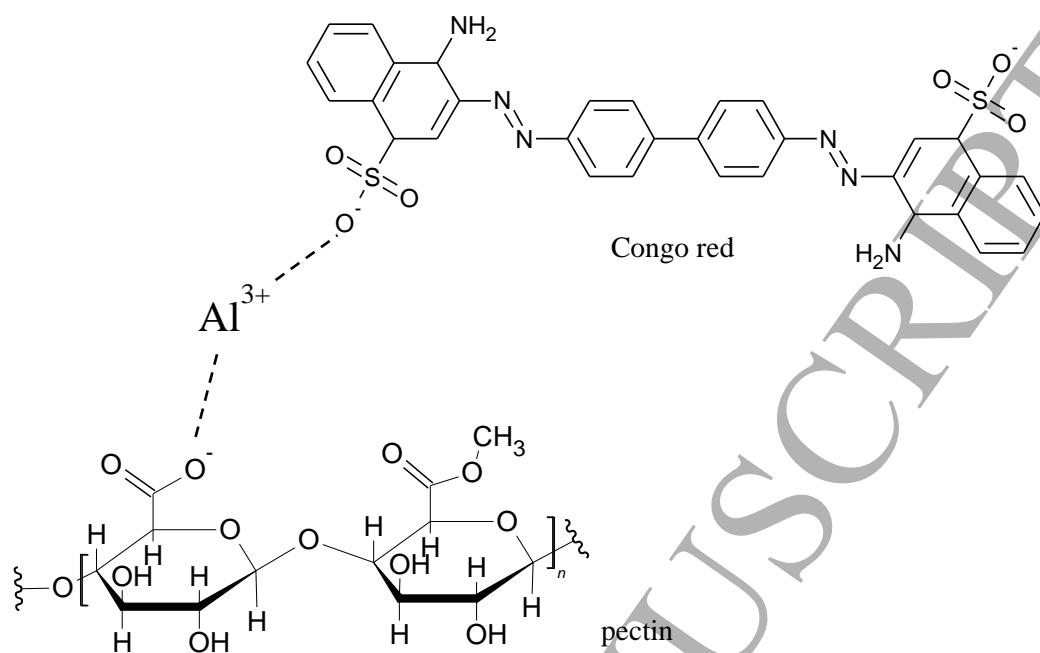
14 **Figure 4.** Observed flocs formation with 15 mg/L pectin (a) and without pectin (b) at pH 6.
15 [alum] = 30 mg/L. [Congo red] = 50 mg/L.
16
17

18 The results obtained in this study are compared with other coagulants for Congo red
19 removal, presented in Table 3. It can be observed that the coagulation performance of alum
20 with the aid of pectin could give a comparable coagulation performance with other coagulants
21 reported in the background literature. Furthermore, it can be seen that a smaller alum dosage
22 (30 mg/L) is needed to obtain a 97% removal with the 15 mg/L pectin addition, compared to
23 research conducted by [41] where 100 mg/L alum was required.
24
25
26
27

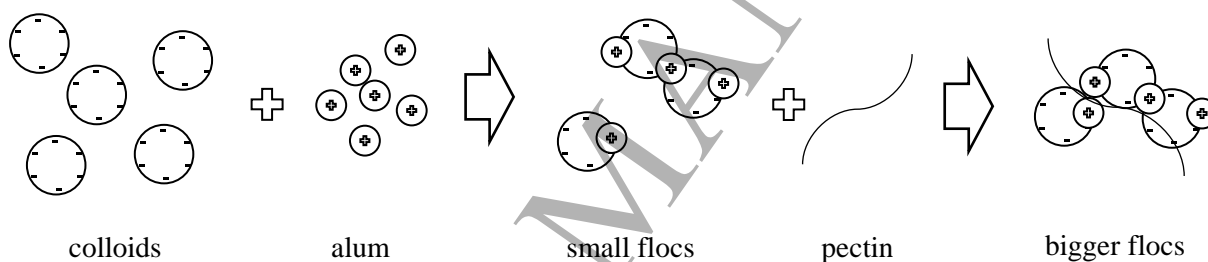
28 **Table 3.** Comparison of various coagulants for Congo red removal

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	<i>A. tetragonus</i>	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]

a.



b.



30
31 **Figure 5.** Possible interaction between Al^{3+} -Congo red and Al^{3+} -pectin (a) and the particle
32 bridging of flocs by pectin (b) (Reused with permission from [40]; Copyright © Elsevier license
33 number 5524610093660)
34

35 36 37 38 39 40 41 42 43 44 45 **4. CONCLUSION**

In this study, low-methoxy pectin has been 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

46 approximately 20% increase of removal compared to alum only. At a higher Congo red
47 concentration, the removal became lower than the one without pectin. This might happen due
48 to an inadequacy of alum at a fixed coagulant dosage with the increase of Congo red
49 concentration. This condition could lead to a competitive interaction of alum-Congo red and
50 alum-pectin, where both Congo red and pectin are negatively charged at the coagulation
51 condition. The results of this research study show that low-methoxy pectin is a potential
52 coagulant-aid that could work synergistically with alum in the coagulation process. Further
53 studies are required to observe the potential of the combination between alum and pectin in
54 other types of wastewaters, real wastewater, and optimization of the variables as well.

55 56 REFERENCES

- 57
- 58 [1] A. K. Verma, R. R. Dash, and P. Bhunia. (2012). "A review on chemical
59 coagulation/flocculation technologies for removal of colour from textile wastewaters".
60 *Journal of Environmental Management*. **93** : 154–168.
61 [10.1016/j.jenvman.2011.09.012](https://doi.org/10.1016/j.jenvman.2011.09.012).
- 62 [2] G. Crini and E. Lichtfouse. (2019). "Advantages and disadvantages of techniques used
63 for wastewater treatment". *Environmental Chemistry Letters*. **17** : 145–155.
64 [10.1007/s10311-018-0785-9](https://doi.org/10.1007/s10311-018-0785-9).
- 65 [3] C.-Y. Yin. (2010). "Emerging usage of plant-based coagulants for water and
66 wastewater treatment". *Process Biochemistry*. **45** : 1437-1444.
67 [10.1016/j.procbio.2010.05.030](https://doi.org/10.1016/j.procbio.2010.05.030).
- 68 [4] H. Kristianto. (2021). "Recent advances on magnetic natural coagulant: a mini review".
69 *Environmental Technology Review*. **10** (1): 254-269.
70 [10.1080/21622515.2021.1986576](https://doi.org/10.1080/21622515.2021.1986576).
- 71 [5] A. Ibrahim, A. Z. Yaser, and J. Lamaming. (2021). "Synthesising tannin-based
72 coagulants for water and wastewater application: A review". *Journal of Environmental
73 Chemical Engineering*. **9** (1): 105007. [10.1016/j.jece.2020.105007](https://doi.org/10.1016/j.jece.2020.105007).
- 74 [6] E. Díaz-Montes. (2022). "Polysaccharides: Sources, Characteristics, Properties, and
75 Their Application in Biodegradable Films". *Polysaccharides*. **3** : 480-501.
76 [10.3390/polysaccharides3030029](https://doi.org/10.3390/polysaccharides3030029).

- 77 [7] C. Y. Teh, T. Y. Wu, and J. C. Juan. (2014). "Optimization of agro-industrial
78 wastewater treatment using unmodified rice starch as a natural coagulant". *Industrial*
79 *Crops and Products*. **56** : 17–26. [10.1016/j.indcrop.2014.02.018](https://doi.org/10.1016/j.indcrop.2014.02.018).
- 80 [8] M. S. Zafar, M. Tausif, M. Mohsin, S. W. Ahmad, and M. Zia-ul-Haq. (2015). "Potato
81 Starch as a Coagulant for Dye Removal from Textile Wastewater". *Water, Air, & Soil*
82 *Pollution*. **226** (8): 244. [10.1007/s11270-015-2499-y](https://doi.org/10.1007/s11270-015-2499-y).
- 83 [9] H. A. Aziz and N. I. M. Sobri. (2015). "Extraction and application of starch-based
84 coagulants from sago trunk for semi-aerobic landfill leachate treatment".
85 *Environmental Science and Pollution Research*. **22** : 16943–16950. [10.1007/s11356-](https://doi.org/10.1007/s11356-015-4895-7)
86 [015-4895-7](https://doi.org/10.1007/s11356-015-4895-7).
- 87 [10] S. Y. Choy, K. N. Prasad, T. Y. Wu, M. E. Raghunandan, and R. N. Ramanan. (2016).
88 "Performance of conventional starches as natural coagulants forturbidity removal".
89 *Ecological Engineering*. **94** : 352-364. [10.1016/j.ecoleng.2016.05.082](https://doi.org/10.1016/j.ecoleng.2016.05.082).
- 90 [11] H. Kristianto, A. Jennifer, A. K. Sugih, and S. Prasetyo. (2020). "Potensi Polisakarida
91 dari Limbah Buah-buahan sebagai Koagulan Alami dalam Pengolahan Air dan Limbah
92 Cair: Review". *Jurnal Rekayasa Proses*. **14** (2): 108-127. [10.22146/jrekpros.57798](https://doi.org/10.22146/jrekpros.57798).
- 93 [12] R. Sanghi, B. Bhattacharya, and V. Singh. (2002). "Cassia angustifolia seed gum as an
94 effective natural coagulant for decolourisation of dye solutions". *Green Chemistry*. **4** :
95 252–254. [10.1039/B200067A](https://doi.org/10.1039/B200067A).
- 96 [13] R. Sanghi, B. Bhattacharya, A. Dixit, and V. Singh. (2006). "*Ipomoea dasysperma*
97 seed gum: An effective natural coagulant for the decolorization of textile dye
98 solutions". *Journal of Environmental Management*. **81** (1): 36-41.
99 [10.1016/j.jenvman.2005.09.015](https://doi.org/10.1016/j.jenvman.2005.09.015).
- 100 [14] K. P. Y. Shak and T. Y. Wu. (2015). "Optimized use of alum together with unmodified
101 *Cassia obtusifolia* seed gum as a coagulant aid in treatment of palm oil mill effluent
102 under natural pH of wastewater". *Industrial Crops and Products*. **76** : 1169–1178.
103 [10.1016/j.indcrop.2015.07.072](https://doi.org/10.1016/j.indcrop.2015.07.072).
- 104 [15] H. Kristianto, S. A. Saraswati, S. Prasetyo, and A. K. Sugih. (2023). "The utilization of
105 galactomannan from spent coffee grounds as a coagulant aid for treatment of synthetic
106 Congo red wastewater". *Environmental Development and Sustainability*. **25** : 5443–
107 5457. [10.1007/s10668-022-02274-x](https://doi.org/10.1007/s10668-022-02274-x).
- 108 [16] K. Q. Lau, M. R. Sabran, and S. R. Shafie. (2021). "Utilization of Vegetable and Fruit
109 By-products as Functional Ingredient and Food". *Frontiers in Nutrition*. **8** : 661693.
110 [10.3389/fnut.2021.661693](https://doi.org/10.3389/fnut.2021.661693).

- 111 [17] S. Y. Choy, K. M. N. Prasad, T. Y. Wu, M. E. Raghunandan, B. Yang, S.-M. Phang,
112 and R. N. Ramanan. (2017). "Isolation, characterization and the potential use of starch
113 from jackfruit seed wastes as a coagulant aid for treatment of turbid water".
114 *Environmental Science and Pollution Research*. **24** : 2876–2889. [10.1007/s11356-016-](https://doi.org/10.1007/s11356-016-8024-z)
115 [8024-z](https://doi.org/10.1007/s11356-016-8024-z).
- 116 [18] M. V. Nsom, E. P. Etape, J. F. Tendo, B. V. Namond, P. T. Chongwain, M. D. Yufanyi,
117 and N. William. (2019). "A Green and Facile Approach for Synthesis of Starch-Pectin
118 Magnetite Nanoparticles and Application by Removal of Methylene Blue from Textile
119 Effluent". *Journal of Nanomaterials*. **2019** : 4576135. [10.1155/2019/4576135](https://doi.org/10.1155/2019/4576135).
- 120 [19] M. C. N. Picot-Allain, B. Ramasawmy, and M. N. Emmambux. (2022). "Extraction,
121 Characterisation, and Application of Pectin from Tropical and Sub-Tropical Fruits: A
122 Review". *Food Reviews International*. **38** (3): [10.1080/87559129.2020.1733008](https://doi.org/10.1080/87559129.2020.1733008).
- 123 [20] M. Moslemi. (2021). "Reviewing the recent advances in application of pectin for
124 technical and health promotion purposes: From laboratory to market". *Carbohydrate*
125 *Polymers*. **254** : 117324. [10.1016/j.carbpol.2020.117324](https://doi.org/10.1016/j.carbpol.2020.117324).
- 126 [21] F. Naqash, F. A. Masoodi, S. A. Rather, S. M. Wani, and A. Gani. (2017). "Emerging
127 concepts in the nutraceutical and functional properties of pectin—A Review".
128 *Carbohydrate Polymers*. **168** : 227-239. [10.1016/j.carbpol.2017.03.058](https://doi.org/10.1016/j.carbpol.2017.03.058).
- 129 [22] H. A. Schols and A. G. J. Voragen. (1996). "Complex pectins:structure elucidation
130 using enzymes". *Progress in Biotechnology*. **14** : 3–19. [10.1016/S0921-](https://doi.org/10.1016/S0921-0423(96)80242-5)
131 [0423\(96\)80242-5](https://doi.org/10.1016/S0921-0423(96)80242-5).
- 132 [23] B. M. Yapo and D. Gnakri. (2014). In "K. G. Ramawat and J.-M. Mérillon (Eds)
133 Polysaccharides: Bioactivity and Biotechnology". Springer, Cham. 1729-1749.
134 [10.1007/978-3-319-16298-0_62](https://doi.org/10.1007/978-3-319-16298-0_62).
- 135 [24] M. Lapointe and B. Barbeau. (2020). "Understanding the roles and characterizing the
136 intrinsic properties of synthetic vs. natural polymers to improve clarification through
137 interparticle Bridging: A review". *Separation and Purification Technology*. **231** :
138 115893. [10.1016/j.seppur.2019.115893](https://doi.org/10.1016/j.seppur.2019.115893).
- 139 [25] S. J. C. d. Rosemond and K. Liber. (2009). "Wastewater treatment polymers identified
140 as the toxic component of a diamond mine effluent". *Environmental Toxicology and*
141 *Chemistry*. **23** (9): 2234-2242. [10.1897/03-609](https://doi.org/10.1897/03-609).
- 142 [26] D. Ibarra-Rodríguez, J. Lizardi-Mendoza, E. A. López-Maldonado, and M. T. Oropeza-
143 Guzmán. (2017). "Capacity of ‘nopal’ pectin as a dual coagulant-flocculant agent for

- heavy metals removal". *Chemical Engineering Journal*. **323** : 19-28.
[10.1016/j.cej.2017.04.087](https://doi.org/10.1016/j.cej.2017.04.087).
- [27] M. Kebaili, S. Djellali, M. Radjai, N. Drouiche, and H. Lounici. (2018). "Valorization of orange industry residues to form a natural coagulant and adsorbent". *Journal of Industrial and Engineering Chemistry*. **64** : 292-299. [10.1016/j.jiec.2018.03.027](https://doi.org/10.1016/j.jiec.2018.03.027).
- [28] F. Shao, J. Xu, J. Zhang, L. Wei, C. Zhao, X. Cheng, C. Lu, and Y. Fu. (2021). "Study on the influencing factors of natural pectin's flocculation: Their sources, modification, and optimization". *Water Environment Research*. **93** (10): 2261-2273.
[10.1002/wer.1598](https://doi.org/10.1002/wer.1598).
- [29] A. A. Cerqueira and M. R. d. C. Marques. (2012). In: " J. S. Gomes (Ed) New Technologies in the Oil and Gas Industry". IntechOpen.
- [30] M. Chethana, L. G. Sorokhaibam, V. M. Bhandari, S. Raja, and V. V. Ranade. (2016). "Green approach to Dye Wastewater Treatment using Biocoagulants". *ACS Sustainable Chemistry and Engineering*. **4** (5): 2495–2507. [10.1021/acssuschemeng.5b01553](https://doi.org/10.1021/acssuschemeng.5b01553).
- [31] S. Zhao, B. Gao, Q. Yue, Y. Wang, Q. Li, H. Dong, and H. Yan. (2014). "Study of Enteromorpha polysaccharides as a new-style coagulant aid in dye wastewater treatment". *Carbohydrate Polymers*. **103** : 179-186. [10.1016/j.carbpol.2013.12.045](https://doi.org/10.1016/j.carbpol.2013.12.045).
- [32] M. H. Zonoozi, M. R. A. Moghaddam, and M. Arami. (2009). "Coagulation/flocculation of dye-containing solutions using polyaluminium chloride and alum". *Water Science & Technology*. **59** : 1343-1351. [10.2166/wst.2009.128](https://doi.org/10.2166/wst.2009.128).
- [33] S.-S. Liu and T.-T. Liang. (2004). "Return sludge employed in enhancement of color removal in the integrally industrial wastewater treatment plant". *Water Research*. **38** (1): 103-110. [10.1016/j.watres.2003.09.006](https://doi.org/10.1016/j.watres.2003.09.006).
- [34] S.-C. Chua, F.-K. Chong, M. A. Malek, M. R. U. Mustafa, N. Ismail, W. Sujarwo, J.-W. Lim, and Y.-C. Ho. (2020). "Optimized Use of Ferric Chloride and Sesbania Seed Gum (SSG) as Sustainable Coagulant Aid for Turbidity Reduction in Drinking Water Treatment". *Sustainability*. **12** (6): 2273. [10.3390/su12062273](https://doi.org/10.3390/su12062273).
- [35] N. A. Awang and H. A. Aziz. (2012). "Hibiscus rosa-sinensis leaf extract as coagulant aid in leachate treatment". *Applied Water Science*. **2** : 293–298. [10.1007/s13201-012-0049-y](https://doi.org/10.1007/s13201-012-0049-y).
- [36] P. W. Wong, T. T. Teng, and N. A. R. N. Norulaini. (2007). "Efficiency of the Coagulation-Flocculation Method for the Treatment of Dye Mixtures Containing Disperse and Reactive Dye". *Water Quality Research Journal*. **42** (1): 54-62.
[10.2166/wqrj.2007.008](https://doi.org/10.2166/wqrj.2007.008).

- 178 [37] Y. A. J. Al-Hamadani, M. S. Yusoff, M. Umar, M. J. K. Bashir, and M. N. Adlan.
179 (2011). "Application of psyllium husk as coagulant and coagulant aid in semi-aerobic
180 landfill leachate treatment". *Journal of Hazardous Materials*. **190** (1-3): 582-587.
181 [10.1016/j.jhazmat.2011.03.087](https://doi.org/10.1016/j.jhazmat.2011.03.087).
- 182 [38] V. B. V. Maciel, C. M. P. Yoshida, S. M. S. S. Pereira, F. M. Goycoolea, and T. T.
183 Franco. (2017). "Electrostatic Self-Assembled Chitosan-Pectin Nano- and
184 Microparticles for Insulin Delivery". *Molecules*. **22** (10): 1707.
185 [10.3390/molecules22101707](https://doi.org/10.3390/molecules22101707).
- 186 [39] J. Kristanda, K. Sandrosa, H. Kristianto, S. Prasetyo, and A. K. Sugih. (2021).
187 "Optimization study of *Leucaena leucocephala* seeds extract as natural coagulant on
188 decolorization of aqueous Congo red solutions". *Arabian Journal for Science and
189 Engineering*. **46** (7): 6275-6286. [10.1007/s13369-020-05008-1](https://doi.org/10.1007/s13369-020-05008-1).
- 190 [40] M. M. Sudirgo, R. A. Surya, H. Kristianto, S. Prasetyo, and A. K. Sugih. (2023).
191 "Application of xanthan gum as coagulant-aid for decolorization of synthetic Congo
192 red wastewater". *Heliyon* **9** (4): E15011. [10.1016/j.heliyon.2023.e15011](https://doi.org/10.1016/j.heliyon.2023.e15011).
- 193 [41] J. Garvasis, A. R. Prasad, K. O. Shamsheera, P. K. Jaseela, and A. Joseph. (2020).
194 "Efficient removal of Congo red from aqueous solutions using phyto-genic aluminum
195 sulfate nano coagulant". *Materials Chemistry and Physics*. **251** : 123040.
196 [10.1016/j.matchemphys.2020.123040](https://doi.org/10.1016/j.matchemphys.2020.123040).
- 197 [42] G. Vijayaraghavan and S. Shanthakumar. (2016). "Performance study on algal alginate
198 as natural coagulant for the removal of Congo red dye". *Desalination and Water
199 Treatment*. **57** : 6384–6392. [10.1080/19443994.2015.1008578](https://doi.org/10.1080/19443994.2015.1008578).
- 200 [43] H. Patel and R. T. Vashi. (2012). "Removal of Congo Red dye from its aqueous solution
201 using natural coagulants". *Journal of Saudi Chemical Society*. **16** : 131–136.
202 [10.1016/j.jscs.2010.12.003](https://doi.org/10.1016/j.jscs.2010.12.003).
- 203