

Synthesis and Characterization of Modified Lignin Flocculants for Congo Red and Methylene Blue Removal

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Abbreviation: Nat. Env. & Poll. Technol.
Website: www.neptjournal.com

Received: 24-09-2025

Revised: 13-12-2025

Accepted: 16-12-2025

Key Words:

Dye removal
 Grafted flocculant
 Modified lignin flocculants
 Congo red
 Methylene blue

Citation for the Paper:

Bailon, M.X., Lim, B.M. and Villadarez, G.M., 2026. Synthesis and characterization of modified lignin flocculants for Congo red and methylene blue removal. *Nature Environment and Pollution Technology*, 25(3), D1872. <https://doi.org/10.46488/NEPT.2026.v25i03.D1872>

Note: From 2025, the journal has adopted the use of Article IDs in citations instead of traditional consecutive page numbers. Each article is now given individual page ranges starting from page 1.



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ABSTRACT

This study focused on synthesizing lignin-based flocculants through grafting sulfonic acid (SA) and [2-(methacryloyloxy)ethyl] trimethylammonium chloride (METAC) using potassium persulfate as the initiator. The resulting flocculants were characterized by particle size, thermal stability, surface morphology, and functional groups. Their dye removal performance was tested with Congo red and methylene blue across various pH levels. FTIR analysis confirmed successful grafting of METAC and SA onto the lignin backbone in lignin-METAC, lignin-SA, and lignin-METAC-SA. Particle size analysis revealed an increase only in lignin-METAC-SA. Modifications led to increased folding and the emergence of raised structures on the flocculants. Thermal analysis showed that, compared to unmodified lignin (degradation at 384°C), all modified lignins exhibited higher maximum degradation temperatures (400–500°C). Modifying lignin improved dye removal efficiency, with lignin-SA demonstrating the highest performance for both dyes. All flocculants performed better against methylene blue at all pH levels, notably lignin-SA (82.9%) and lignin-METAC-SA (82.3%) at pH 10. Higher Congo red removal (46–69%) occurred in more acidic conditions (pH 4). These findings underscore the potential of grafted lignin-based flocculants for dye wastewater treatment.

1. INTRODUCTION

As water scarcity and pollution continue to worsen, advances in wastewater treatment and management are essential (Libralato et al. 2012). Wastewater is processed through chemical, biological, physical, or combined methods to reduce contaminants and achieve the required water quality standards (Englande et al. 2015). The textile dyeing and finishing industry ranks as the second-largest polluter of clean water, after agriculture. Around 8,000 chemicals are involved in textile manufacturing, including dyeing and printing, many of which pose known environmental and health risks (Kant 2012, Yaseen & Scholz 2019). Consequently, dye wastewater must be treated prior to discharge into surface water bodies to mitigate its environmental impact (Solayman et al. 2023).

Various treatment methods have been explored and developed for dye removal from wastewater. Nonetheless, some approaches fail to meaningfully reduce contaminant levels, and practical methods often face limitations such as high costs and the risk of secondary pollution. For example, chemical precipitation is a straightforward and effective technique for high pollutant concentrations but involves substantial chemical usage and generates significant sludge. Another common method, adsorption or filtration, is versatile and effective across a broad

spectrum of contaminants; however, it is constrained by the expense of filtration materials, ongoing maintenance to prevent saturation and clogging, and the need for regeneration of the media (Crini & Lichtfouse 2019). An optimal treatment process would substantially lower dye concentrations in wastewater while avoiding the creation of potentially hazardous by-products (Katheresan et al. 2018).

Flocculation is a well-established method for removing dyes. This process involves adding coagulants, which destabilize particle forces, and flocculants, which facilitate the aggregation of contaminants into larger flocs through bridging, thereby separating solids from liquids (Arockiam JeyaSundar et al. 2020). Unlike some other methods, flocculation poses minimal negative impacts and is frequently used to decrease water turbidity by eliminating suspended solids, dissolved pollutants, and emulsified oils (Das et al. 2021). The process can be optimized to lower chemical consumption, sludge generation, and costs by employing direct flocculation, which involves adding only flocculants to wastewater. For example, polyelectrolytes-polymers with either a cationic or anionic charge, such as chitosan, are commonly used as flocculants (Chatsungnoen & Chisti 2019).

Synthetic polymer flocculants, such as polyacrylamide, have been used in different industries because of their high molecular weight and exceptional performance in contaminant removal. However, they are non-biodegradable and can pose hazards to living organisms (Siti Aisyah et al. 2014). Natural flocculants have attracted attention in several studies because of their comparable performance to synthetic flocculants (Ajao et al. 2018), safer use, and ability to produce organic sludge that is biodegradable and reusable as fertilizer in agriculture (Fauzani et al. 2021, Zaman et al. 2020). One of the common biopolymers used as a flocculant is plant-derived lignin because of its natural abundance, biodegradability, and presence of functional groups that can effectively interact with contaminants during the flocculation process (Suryawanshi et al. 2025).

Flocculation involves the chemical destabilization of particles by overcoming their repulsive forces through complex bridging mechanisms (Gregory & O'Melia, 1989). Lignin can be chemically modified to change its surface charge by grafting various functional groups onto its backbone (Lee et al. 2014, Wang et al. 2008). For example, Wang et al. (2018) grafted [2-(methacryloyloxy)ethyl] trimethylammonium chloride (METAC) onto lignin to produce a more cationic flocculant. This lignin-METAC flocculant demonstrated effective removal of anionic dyes, such as Remazol Black B and Remazol Brilliant Orange, via bridging and charge neutralization mechanisms.

Alternatively, lignin can be rendered more anionic by attaching negative groups, like sulfone or sulfonate groups, which increase its negative charge density (Aro & Fatehi 2017, Gao et al. 2019). Such modifications have proven effective for removing cationic dyes like methylene blue (Rahmawati et al. 2021). Additionally, lignin can be engineered to create amphoteric flocculants capable of removing both cationic and anionic dyes (Yue et al. 2005).

Wu et al. (2023) developed an amphiphilic lignin flocculant by grafting acrylamide and 3-chloro-2-hydroxypropyltrimethylammonium chloride onto the kraft lignin backbone, demonstrating its effectiveness in treating high-turbidity wastewater. Eraghi Kazzaz and Fatehi (2020) examined the cationization of lignin with (3-bromopropyl) trimethylammonium bromide and (5-bromopentyl) trimethylammonium bromide, combined with polymerization using 3-sulfopropyl methacrylate, though its impact on flocculation performance remains unstudied. Additionally, the Mannich reaction applied to acrylamide-modified lignosulfonate with amine and aldehyde yielded the amphoteric lignin flocculant LSDC, which proved effective in decolorizing various dyes by up to 80% under appropriate conditions (Yue et al. 2005). Collectively, these studies highlight the potential to enhance lignin's flocculation and contaminant removal capabilities through the incorporation of both cationic and anionic groups.

To the best of our knowledge, no previous research has reported the grafting of anionic sulfonic acid and cationic METAC groups onto lignin, nor have they been evaluated for dye removal effectiveness. In this study, three lignin-based flocculants were synthesized by grafting METAC and sulfonic acid (SA) groups onto the lignin backbone using potassium persulfate as an initiator. These synthesized flocculants were characterized and tested for their ability to remove dyes, specifically methylene blue and Congo red. All the lignin-based flocculants demonstrated effectiveness in removing both dyes, indicating their promising potential for dye wastewater treatment.

2. MATERIALS AND METHODS

2.1. Materials

Alkali lignin, [2-(methacryloyloxy)ethyl] trimethylammonium chloride (METAC) at 75% weight in water, and potassium persulfate ($K_2S_2O_8$) with a purity of at least 99% were obtained from Merck Sigma-Aldrich. Linear alkylbenzene sulfonic acid (LABSA) was sourced from XERN Chemicals in the Philippines. All reagents were used as received, without further purification.

2.2. Synthesis of Modified Lignin Flocculants

Three variants of modified lignin-based flocculants were synthesized through grafting METAC and sulfonic acid (SA) onto the lignin backbone. The preparation followed methods outlined by Liu et al. (2018) and Moore et al. (2021), with specific modifications. Briefly, alkali lignin was dissolved in distilled water at a 1:20 ratio of solute to solvent, and the mixture was stirred at 400 rpm until fully dissolved. A 0.1 mol.L⁻¹ NaOH solution was gradually added to adjust the pH to 11. The solution was then heated to 70°C for 30 minutes, after which the pH was readjusted to 4 using 0.1 M H₂SO₄. These pH adjustments optimized solubility and reaction conditions. The mixture was maintained at 75°C in a water bath until the end of the synthesis and purged with nitrogen for 30 minutes. Subsequently, 0.015 g of potassium persulfate per gram of lignin was introduced as a free radical initiator while stirring at 400 rpm. Depending on the desired flocculant type, METAC, SA, or both were added dropwise in a 1:2 solute-to-modifier weight ratio, as detailed in Table 1. The reaction mixture was stirred for 2.5 hours to ensure complete grafting. After completion, the solution was mixed with 80% ethanol (v/v) to precipitate the flocculants. The precipitate was separated, washed twice with 80% ethanol, and then dried at 100°C.

2.3. Sample Characterization

This study conducted a functional group analysis of various flocculants utilizing a Cary 360 FTIR Spectrometer (Agilent Technologies) at the Philippine Science High School – Central Luzon Campus. IR spectra were recorded by measuring the percent transmittance across a range of 650 to 4000 cm⁻¹. The particle sizes of different lignin-based flocculants were examined through optical microscopy at the Department of Science and Technology – Industrial Technology Development Institute (DOST-ITDI), Material Science Division. Images of each sample were captured and analyzed to determine particle size distribution. Optical microscopy was chosen because the samples were anticipated to fall within the 50–200 micron range, which exceeds the optimal detection range of Dynamic Light Scattering (DLS). The surface morphologies of the flocculants were

Table 1: Mass ratios for the lignin and modifier used in the preparation of the different flocculants.

Type	Lignin to METAC to SA Mass Ratio		
	Lignin	METAC	SA
Unmodified Lignin	1	0	0
Lignin-METAC	1	2	0
Lignin-SA	1	0	2
Lignin-METAC-SA	1	2	2

observed using a HITACHI TM4000Plus Scanning Electron Microscope (SEM) at the Philippine Science High School – Ilocos Region Campus, at magnifications between 800× and 1200×. The thermal stability was assessed with a Netzsch STA 2500 Regulus Simultaneous Thermal Analyzer, heating samples from 20°C to 600°C at a ramp rate of 40 K.min⁻¹.

2.4. Dye Removal Performance Against Congo Red and Methylene Blue

A dye removal experiment was performed following the protocols of Wang et al. (2018) and Wu et al. (2022), with some modifications. Various flocculants were assessed for their effectiveness in removing Congo red and methylene blue dyes at different pH levels (pH 4, 7, and 10). These dyes were chosen due to their differing charges, with Congo red being an anionic dye and methylene blue a cationic dye. The flocculants were applied by adding 100 mg/L of each to 50 mL of artificial dye wastewater, which had a concentration of 10 mg/L, in centrifuge tubes. The mixtures were shaken for 2 hours on a linear shaker and then left to settle for an additional 2 hours. The supernatant was filtered through a 0.45 µm PTFE membrane, and the absorbance was measured using a Shimadzu UV-10008 UV-Visible spectrophotometer at 497 nm for Congo red and 664 nm for methylene blue. The dye removal efficiency of the flocculants was compared to that of RAFLOC 28 (a cationic polymer flocculant) and ARFLOC 12 (an anionic polymer flocculant).

2.5. Statistical Analysis

A One-way ANOVA was conducted at a 95% confidence level to assess the impact of different flocculants on the reduction of the two dyes' concentrations. Tukey's HSD post hoc test was then applied to determine significant differences in the performance of the various flocculants, including the positive controls.

3. RESULTS AND DISCUSSION

3.1. FTIR Analysis

Fig. 1 shows the FT-IR spectra of unmodified lignin and the different synthesized lignin-based flocculants (Lignin-METAC, Lignin-SA, and Lignin-METAC-SA). Characteristic peaks for hydroxyl moieties (-OH: 3400 cm⁻¹) and methyl and methylene vibrations (alkyl -CH stretch: 2900 cm⁻¹) were present in all samples, suggesting the presence of many hydroxyl and methylene groups, which are highly associated with lignin (Guo et al. 2018). The different flocculants also exhibited prominent peaks at 1650 cm⁻¹ and 1200 cm⁻¹ associated with -C=C- of the characteristic conjugated alkene of lignin and -CO stretching

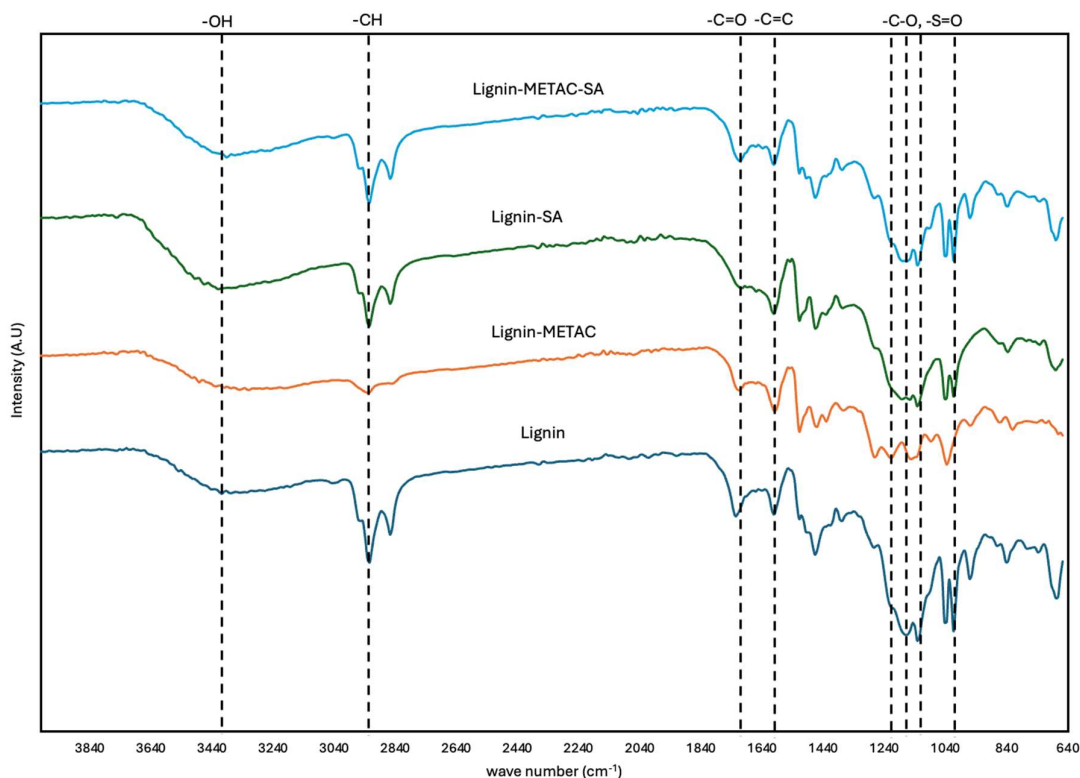


Fig. 1: FTIR spectra of unmodified lignin and synthesized modified lignin flocculants.

vibrations, respectively. Characteristic peaks for -C=O stretching (1750 cm^{-1}) and stretching vibration for $\text{N}^+(\text{CH}_3)_3$ (960 cm^{-1}) were observed for Lignin-METAC and Lignin-METAC-SA, suggesting the successful incorporation of METAC in the structure of lignin (Guo et al. 2018). Moore et al. (2021) described that the mechanism for modification is through METAC grafting through the hydroxyl groups. This eventually translates to an apparent reduction in the -OH peak at approximately 3300 cm^{-1} . Furthermore, the peaks for the -C=O stretch around 1750 cm^{-1} were more prominent in Lignin-METAC and Lignin-METAC-SA than in Lignin-SA, which is expected as METAC contains a carbonyl group. In contrast, peaks around $1020\text{-}1220\text{ cm}^{-1}$ associated with -S=O stretching due to sulfonate and sulfonic acid groups were observable in Lignin, Lignin-SA, and Lignin-METAC-SA, but highly diminished in Lignin-METAC (Karpukhina et al. 2023). These results confirm the successful grafting of METAC and sulfonic acid structures to the lignin backbone.

3.2. Particle Size Analysis

Particle size analysis of various flocculants was performed using optical microscopy (Fig. 2). Results indicated that lignin, lignin-METAC, and lignin-SA have comparable particle sizes, approximately 1.5 to $1.7\text{ }\mu\text{m}$. In contrast, lignin-METAC-SA exhibited significantly larger particles,

with some reaching a Feret diameter of $150\text{ }\mu\text{m}$. The addition of METAC and SA to lignin likely alters the inter- and intramolecular forces, potentially causing structural folding and interlinking among lignin molecules. This interaction may lead to an increase in particle size for lignin-METAC-SA. Specifically, electrostatic interactions between opposite charges and the possible formation of ionic bonds within lignin-METAC-SA could account for its larger particle dimensions compared to lignin-SA and lignin-METAC (Lekniute et al. 2013).

3.3. Scanning Electron Microscopy

Scanning electron microscopy images (Fig. 3) revealed differences among the flocculants. Unmodified lignin displayed uniform spherical structures with mounds and grooves, while lignin-SA and lignin-METAC showed more porous microstructures. The modified lignin flocculants also exhibited increased folds and raised features. The presence of crazing and cracks indicates the formation of a plastic phase in the flocculants (Moustaqim et al. 2018). These surface morphology changes may result from disrupted hydrogen bonding in alkali lignin during modification, leading to the collapse of its crystal structure and subsequent surface alterations (Li et al. 2024). Confirmation of these

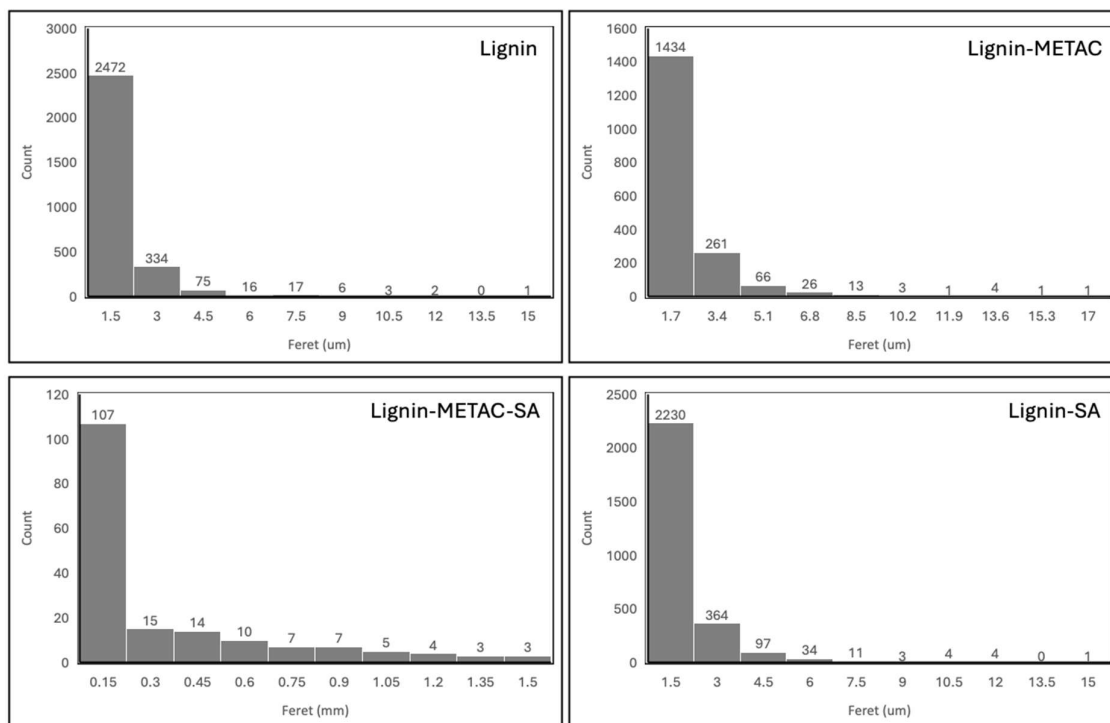


Fig. 2: Particle size distribution of pure lignin and modified lignin flocculants.

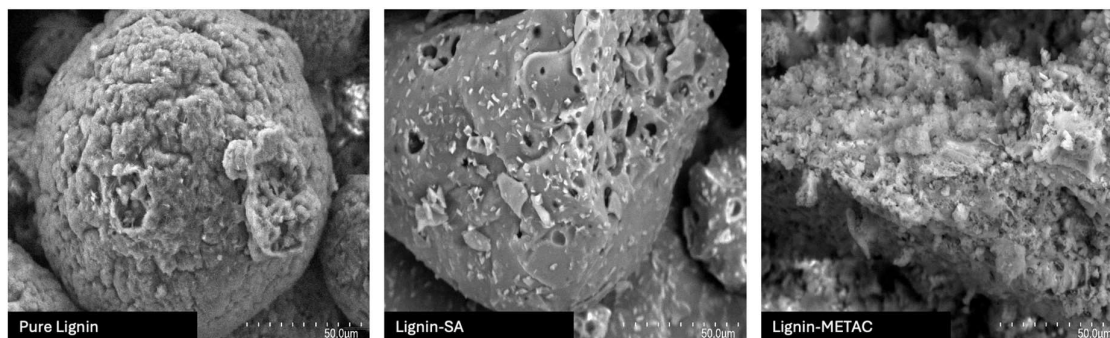


Fig. 3: SEM image of lignin-based flocculants at 800x magnification.

effects could be achieved through molecular weight and zeta potential analyses, which were not conducted in this study.

3.4. Thermogravimetry Analysis

The thermal stability of each flocculant was evaluated using a simultaneous thermal analyzer. The TGA profiles (Fig. 4) exhibit differences in the thermal behavior of the various flocculants, which is expected due to the incorporation of new functional groups into the lignin structure. Overall, the patterns in the TGA and DTG curves were similar across all samples, with the DTG traces showing distinct peaks around 70–150°C and 250–500°C. These peaks correspond to apparent inflections in the thermograms of the different

samples. Both exothermic and endothermic peaks are evident in the DTG curves, with the maximum exothermic peaks occurring at higher temperatures in the modified lignin flocculants compared to unmodified lignin. The DTG curves displayed three to four peak temperatures of maximum weight loss (T_p) for each flocculant, as summarized in Table 2. Additionally, all flocculants demonstrated a slower decomposition rate at temperatures exceeding 500°C.

The first peak (T_{p1}) in the DTG at approximately 73–162°C is attributed to the loss of moisture, as much of the mass loss occurred around the vaporization point of water. The flocculants exhibited differences in the 2nd to 4th peak temperatures owing to structural differences. In

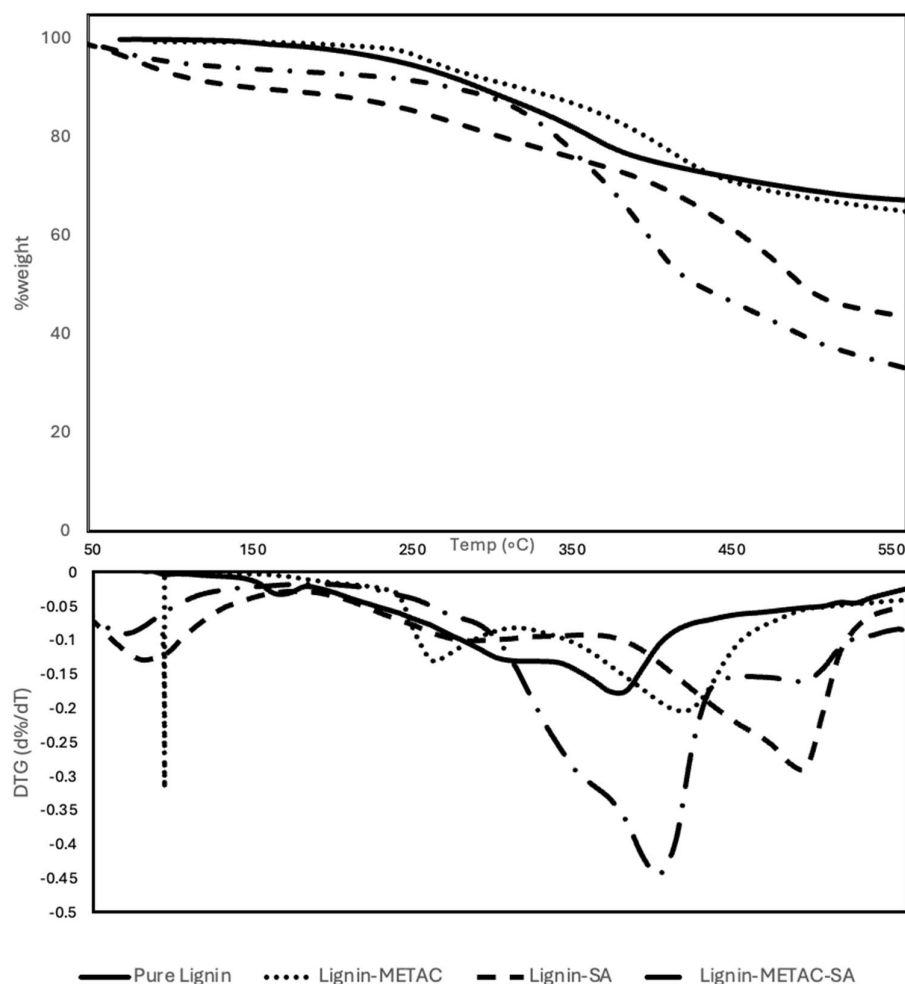


Fig. 4: TGA and DTG graphs for pure lignin, lignin-METAC, lignin-SA, and lignin-METAC-SA flocculants.

the case of unmodified lignin, unsaturated chains, methane, carbon dioxide, and carbon monoxide are formed after dehydration. The process continues with the breaking of the methyl-aryl ether bonds at approximately 300°C and phenol ring decomposition at approximately 400°C (Moore et al. 2021, Sahoo et al. 2011). The mass loss at 304°C is also due to the cleavage of the b-O-4 bonds of carbohydrate molecules (Chu et al. 2013, Kim et al. 2013, Moustaqim et al. 2018).

In the case of lignin-METAC, T_{P2} occurred at a lower temperature (261.9°C), which may be due to the decomposition of the quaternary ammonia group. Simultaneously, T_{P3} is attributed to the decomposition of the Lignin-METAC backbone (Moore et al. 2021). The same decomposition pattern was observed for Lignin-METAC-SA, but at a higher temperature, owing to the incorporation of both METAC and SA groups. T_{P3} for all modified flocculants occurred at temperatures between 400 and 500°C, which

is due to the saturation reaction of the aromatic ring, C-C bond breakage, and the release of water, carbon dioxide, and carbon monoxide by-products during the rearrangement and removal of functional groups (Jakab et al. 1991, Wibowo & Park 2023).

3.5. Dye Removal Performance

The influence of pH on the flocculating performance of various flocculants was examined using Congo Red dye, with

Table 2: Peak weight loss temperature (T_p) of pure lignin, lignin-METAC, lignin-SA, and lignin-METAC-SA.

	Peak Temperatures [°C]			
	T_{P1}	T_{P2}	T_{P3}	T_{P4}
Unmodified lignin	162	302	384	-
lignin-METAC	94.7	261.9	425	-
lignin-SA	88.06	281	496.8	-
lignin-METAC-SA	72.6	363	404.6	497.1

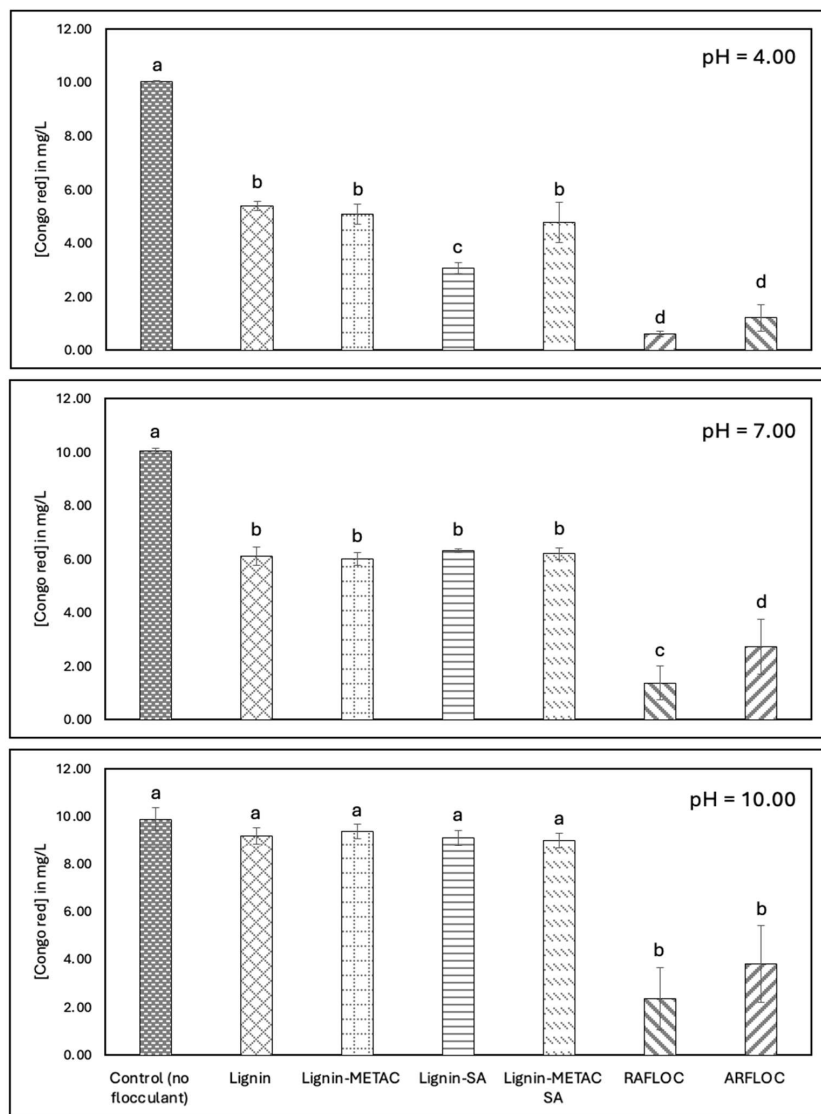


Fig. 5: Effect of pH on the dye removal performance of unmodified lignin, Lignin-METAC, Lignin-SA, and Lignin-METAC-SA flocculants against Congo red dye. Letters above the bars represent a significant difference among treatment groups based on ANOVA ($p < 0.05$) and Tukey's HSD test. Similar letters mean no significant difference between treatments. Error bars indicate standard deviation ($n = 3$).

results summarized in Fig. 5. All lignin-based flocculants effectively removed Congo Red at pH 4 and 7. Under acidic conditions, unmodified lignin, lignin-METAC, and lignin-METAC-SA exhibited similar reduction efficiencies, ranging from approximately 46% to 52%. Among the synthesized flocculants, lignin-SA achieved the highest performance, with an average reduction efficiency of 69.4%. At neutral pH, all lignin-based flocculants demonstrated comparable performance, with reduction rates between 37% and 40%. No significant dye removal was observed at pH 10 for any lignin-based flocculant. Across all pH levels, the positive control groups, RAFLOC and ARFLOC, showed superior

performance, with average reduction efficiencies at pH 4, 7, and 10 of approximately 88-94%, 73-86%, and 61-76%, respectively.

Both RAFLOC and ARFLOC positive controls showed significantly higher reduction compared to lignin-based flocculants. This was expected because both are commercially used flocculants for treating industrial wastewater. Except at pH 10, all lignin flocculants exhibited significant dye removal activity. The isoelectric point of Congo Red is pH 3 (Litefti et al. 2019). Thus, it has been observed to ionize into a more positive charge ion below pH 3 and exhibit a more negative charge at values above pH 5.3 (Alarifi et al.

2021). It is expected that the anionic flocculant Lignin-SA will perform the best because of its ability to neutralize and destabilize dye contaminants, reducing electric repulsion, which will lead to efficient aggregation of flocs (Gheraout & Gheraout 2012). In addition, sulfonation generally increases the solubility of polymers in water, which increases the ability of the flocculant to adsorb onto the dye particles and bridge them together (Lufrano et al. 2013).

In contrast, lignin-METAC, with its cationic nature, tends to undergo flocculation via polymer bridging. This relies on several additional factors, including the agitation of the

particles and flexibility of the polymer, as well as the surface charges, branches, and functional groups that can serve as interaction points, which may lead to weaker flocculation performance (Hogg 2013). This may also explain why lignin-METAC-SA performed better than lignin-METAC but worse than lignin-SA, as the amphoteric nature may have diminished some of its ability to neutralize dye contaminants. However, its polymer-bridging capability remains owing to the polymeric nature of the grafted flocculants. Unmodified lignin also performed flocculation with an efficiency comparable to that of lignin-METAC. This is likely due to

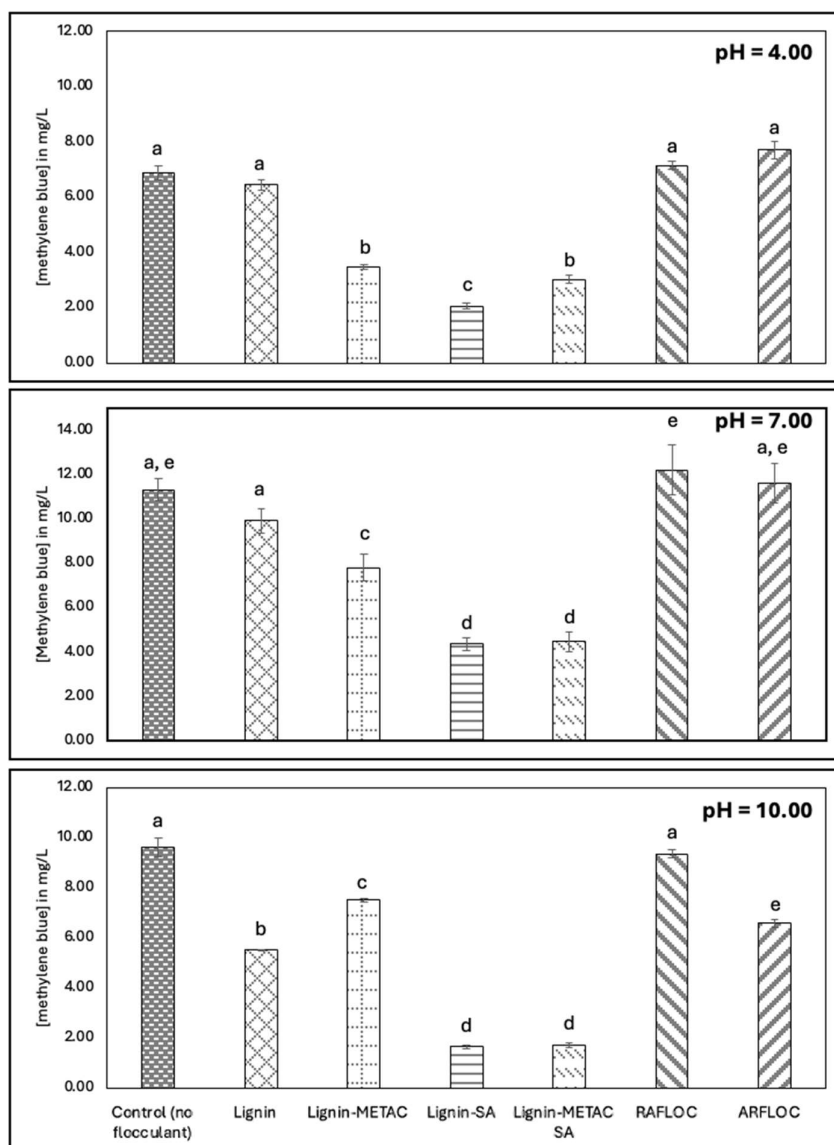


Fig. 6: Effect of pH on the dye removal performance of unmodified lignin, Lignin-METAC, Lignin-SA, and Lignin-METAC-SA flocculants against methylene blue dye. Letters above the bars represent a significant difference among treatment groups based on ANOVA ($p < 0.05$) and Tukey's HSD test. Similar letters mean no significant difference between treatments. Error bars indicate standard deviation ($n = 3$).

the mechanism utilized by these two flocculants, except that Lignin-METAC had additional functional groups that could still interact with the contaminants through intermolecular forces, increasing the likelihood of bridging. At pH 7 and 10, RAFLOC still performed best for dye removal, which can be attributed to its cationic nature, which can effectively neutralize Congo red, which is anionic at these pH levels, through charge neutralization. Similar to other polyacrylamide flocculants, the sweeping effects and enmeshment of RAFLOC and ARFLOC allowed for the removal of the Congo red dye through possible adsorption and bridging mechanisms (Khan et al. 2020).

The dye removal performance of the different flocculants was also tested using the methylene blue dye. Fig. 6 shows that at all pH levels, the unmodified lignin and all modified lignin flocculants showed significant dye removal performance. At pH 4.00, the Lignin-METAC, Lignin-SA, and Lignin-METAC-SA flocculants showed high average reduction efficiencies of 49.63%, 79.15%, and 56.04%, respectively. Unmodified lignin and the two positive controls did not show a significant reduction at this pH. Similar to that at pH 4.00, no significant reduction in dye concentration was observed for unmodified lignin and the two positive controls. The highest average reduction efficiencies were observed for Lignin-SA (61.5%) and Lignin-METAC-SA (60.7%). Lignin-METAC also exhibited a significant dye removal performance of 31.1%. At pH 10, all flocculants except the RAFLOC control showed significant flocculation performance. Lignin-METAC showed the lowest average reduction at 22%, followed by ARFLOC (31.7%) and unmodified lignin (42.9%). Lignin-SA and lignin-METAC-SA showed the highest average reduction efficiencies at 82.9% and 82.3%, respectively.

The observed trends for methylene blue and Congo red can be explained by their charge differences, with methylene blue being a cationic dye. All lignin-based flocculants demonstrated more effective dye removal than the positive controls, RAFLOC and ARFLOC. The results of the methylene blue flocculation test underscore the beneficial effect of grafting cationic and anionic groups onto the lignin backbone, which enhances its flocculating performance. Unmodified lignin showed minimal dye reduction at pH 4 and 7. Lignin modified with significant sulfonic acid groups (lignin-SA and lignin-METAC-SA) carries more negative charges, enabling effective neutralization of the positively charged methylene blue. This aligns with previous research indicating high methylene blue removal efficiency with lignosulfonate (Sotelo et al. 2024, Zhang et al. 2020). For lignin-METAC, polymer bridging facilitated by intermolecular forces was likely the main mechanism,

resulting in weaker flocculation. Lignin-METAC-SA appeared to utilize a combination of these mechanisms.

As the solution became more alkaline, lignin-based flocculants containing sulfonic acid groups showed a marked improvement in dye reduction efficiency. The basic environment likely neutralized the H^+ ions in the sulfonic acid groups, leading to the formation of negatively charged SO_3^- ions after dissociation. This process enhances the flocculants' ability to neutralize the positively charged methylene blue dye. Conversely, the lower reduction efficiency observed with Lignin-METAC may result from the neutralization of its quaternary ammonium group's positive charge.

3.6. Limitations and Future Work

While the current study demonstrates the effectiveness of modified lignin in removing Congo red and methylene blue dyes, a significant limitation is the lack of thorough characterization of the synthesized flocculants. Without detailed analysis, it is difficult to elucidate the precise mechanisms of dye removal. For instance, confirming whether electrostatic charge neutralization, bridging, or sweeping effects are involved requires additional investigations. Determining molecular weight is essential to understanding polymer chain length and its role in interparticle bridging. Tests for solubility and viscosity are necessary to assess the polymer's hydrodynamic volume and conformational stability in dye solutions. Zeta potential analysis is also needed to track surface charge changes and identify the point of zero charge for the flocculants. Until these parameters are measured, the exact mechanisms by which the different flocculants remove dyes cannot be conclusively determined.

4. CONCLUSIONS

Lignin-based flocculants modified with different functional groups were successfully synthesized by grafting METAC and sulfonic acid groups onto the lignin backbone. These modifications enhanced surface functionalities and introduced folding and protrusion structures, which likely increased interactions with target dye molecules. The modified lignins demonstrated improved dye removal capabilities for both Congo red and methylene blue across all pH levels, with Lignin-SA and Lignin-METAC-SA achieving removal efficiencies exceeding 80%. The efficacy of the lignin-based flocculants was influenced by the solution pH: higher removal rates for Congo red occurred under more acidic conditions, while methylene blue removal was more effective in alkaline environments. Although the modified lignins were effective in removing both dyes, they were

less efficient than commercial flocculants for Congo red in neutral and alkaline pH conditions. To deepen understanding of the removal mechanism, further characterization, such as molecular weight and zeta potential analysis, could be performed. Additionally, testing flocculating performance across different temperatures and dosing rates is recommended.

5. ACKNOWLEDGMENTS

The authors wish to acknowledge the support and assistance provided by the Philippine Science High School – Central Luzon Campus towards the completion of this research project. The authors also want to acknowledge the support of the Philippine Science High School, Ilocos Region Campus and the DOST, Industrial Technology Development Institute for assisting in the conduct of scanning electron microscopy and particle size analysis, respectively. This research was funded by the Department of Science and Technology, Republic of the Philippines, Philippine Council for Industry, Energy, and Emerging Technology Research and Development under the Young Innovators Program [PCIEERD Project No.: 1212039].

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