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Effectiveness Analysis of Natural and Synthetic Coagulants in Domestic Wastewater Treatment: Coagulation–Flocculation Mechanism and Water Chemical Characterization Study

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ABSTRACT

Domestic wastewater treatment using natural coagulants is gaining attention as a sustainable alternative to synthetic chemicals. This study compared the effectiveness of three natural coagulants (*Moringa oleifera* seed extract, *Phaseolus vulgaris* saline extract, and chitosan) against two synthetic coagulants (polyaluminum chloride, PAC, and alum) for treating real, moderately high-strength domestic wastewater from Yogyakarta, Indonesia. Jar tests were performed in triplicate, measuring turbidity, total suspended solids (TSS), chemical oxygen demand (COD), and pH. Results showed that PAC at 14 mg/L achieved the highest turbidity removal (94.2 ± 2.1%), followed by alum (89.5 ± 3.4%) and *M. oleifera* at a much higher dosage of 4000 mg/L (86.3 ± 4.2%). *P. vulgaris* extract (0.4 mL/L) removed 79.1 ± 5.0% of turbidity, while chitosan (50 mg/L) removed 64.2 ± 6.1%. For COD removal, PAC reached 63.5 ± 3.2%, chitosan 58.3 ± 4.7%, and *M. oleifera* 55.7 ± 4.5%. A hybrid formulation combining 3.5 mg/L PAC and 0.3 mL/L *P. vulgaris* extract achieved 91.8% turbidity removal, nearly matching PAC alone while using 75% less synthetic chemical. We conclude that although PAC remains the most reliable coagulant for high-strength wastewater, natural coagulants especially in hybrid forms offer a viable, low-toxicity option for decentralized treatment applications.

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1. Introduction

Domestic wastewater from household activities is a major source of water pollution in many Indonesian cities [1]. Coagulation and flocculation are key steps in wastewater treatment, as they destabilize suspended particles and dissolved organic matter to form larger flocs that settle out [2,3]. For decades, synthetic chemicals such as alum and polyaluminum chloride (PAC) have been widely used because they are cheap and effective. However, residual aluminum in treated water and the voluminous, chemically laden sludge generated from synthetic coagulants have raised concerns about potential neurotoxicity, ecotoxicity, and long-term environmental persistence [3,4,5].

These drawbacks have pushed researchers to look for greener alternatives. Natural coagulants derived from plants like *Moringa oleifera* seeds, common beans, or chitosan from crustacean shells are biodegradable, non-toxic, and produce less sludge [6,7]. *Moringa oleifera* seed extract, in particular, has been studied extensively, as its cationic proteins can neutralize negatively charged particles through adsorption and charge neutralization [8,9]. Chitosan also shows promise for removing organic matter and microorganisms [10,11]. Meanwhile, seed extracts from common bean (*Phaseolus vulgaris*) have demonstrated coagulation activity, especially when extracted with saline solutions [9].

But natural coagulants are not without problems. *Moringa oleifera* often requires very high dosages (up to several thousand mg/L) and long settling times [8]. Some studies have reported that excessive doses of plant-based coagulants can increase effluent BOD and COD due to the release of soluble organic compounds from the seeds themselves [12]. Most previous research has used synthetic or lightly contaminated water, leaving a gap in our understanding of how these natural materials perform on real, moderately high-strength domestic wastewater. To our knowledge, few studies have directly compared natural, synthetic, and hybrid coagulants using real, moderately high-strength domestic wastewater under identical operating conditions. We hypothesized that a hybrid formulation combining a low dose of PAC with a natural coagulant could achieve comparable pollutant removal to PAC alone while reducing synthetic chemical use. The main aim of this study was to directly compare the performance of three natural coagulants (*Moringa oleifera*, *Phaseolus vulgaris*, and chitosan) against two synthetic coagulants (PAC and alum) using actual domestic wastewater from Yogyakarta, and to explore whether a simple hybrid mixture could offer a balance between effectiveness and environmental safety.

2. Materials and Method

Wastewater Sampling and Initial Characterization

Domestic wastewater was collected from a communal collection point in Jetis District, Yogyakarta, in September 2024. The sampling site was selected because it represents a densely populated residential area with typical domestic wastewater characteristics, making it suitable for evaluating coagulation performance under realistic conditions. Multiple grab samples were collected from the sampling location and composited to minimize temporal variability. Samples were transported under refrigerated conditions and analyzed within 48 h to preserve wastewater characteristics. Initial characterization was performed in triplicate using standard methods: pH (electrometric method), turbidity (nephelometric method), total suspended solids (TSS; APHA 2540 D), chemical oxygen demand (COD; closed reflux colorimetric method), and 5-day biochemical oxygen demand (BOD_5; APHA 5210 B). The raw wastewater exhibited the following characteristics (mean ± SD): pH 7.1 ± 0.2, turbidity 238 ± 18 NTU, TSS 298 ± 32 mg/L, COD 465 ± 41 mg/L, BOD_5 208 ± 22 mg/L. These values exceed the Indonesian domestic wastewater quality standard (Ministry of Environment and Forestry Regulation No. P.68/2016).

Coagulant Preparation

Natural Coagulants

Only mature and undamaged Moringa oleifera and Phaseolus vulgaris seeds were selected to ensure uniform physicochemical properties and consistent extraction efficiency throughout the experiments, and the kernels were dried at 60 °C for 24 h before being ground into a fine powder. For each experiment, 10 g of powder was mixed with 100 mL of distilled water, stirred at 500 rpm for 15 min, and subsequently filtered. The protein concentration determined using the Lowry method was 2.8 ± 0.3 g/L. Meanwhile, dried Phaseolus vulgaris (common bean) seeds were ground using the same procedure, after which 10 g of powder was mixed with 100 mL of 0.5 mol/L NaCl solution, stirred, and filtered following the method described by Pavlović et al. [9]. In addition, the chitosan solution was prepared by dissolving Chitosan (medium molecular weight, >85% deacetylated, Sigma-Aldrich) at a concentration of 1% (w/v) in 1% (v/v) acetic acid according to the procedure reported by de Paula et al. [10].

Synthetic Coagulants

Commercial-grade Aluminum sulfate (alum) was prepared as a 10 g/L stock solution prior to use. Meanwhile, Polyaluminum chloride (PAC) was obtained as a commercial liquid containing 10% Al_2O_3 and diluted as required for the experiments.

Jar Test Procedure

The standard jar test method was employed because it is widely accepted for evaluating coagulation–flocculation efficiency and determining optimum coagulant dosage under controlled laboratory conditions. Each beaker contained 500 mL of wastewater. The tested dosage ranges were: alum 10–60 mg/L, PAC 5–30 mg/L, *Moringa oleifera* 500–6000 mg/L (powder equivalent), *Phaseolus vulgaris* extract 0.1–2.0 mL/L, and chitosan 10–100 mg/L. The standard procedure consisted of rapid mixing at 130 rpm for 1.5 min, slow mixing at 30 rpm for 20 min, and settling for 30 min. Supernatant samples were withdrawn from 2 cm below the water surface. Each condition was run in triplicate (n = 3). The optimum dosage was determined based on the highest removal efficiency for turbidity and COD while maintaining stable pH, rapid floc formation, and practical chemical consumption.

Hybrid Coagulation Test

A hybrid formulation combining 25% of the PAC optimum dose (3.5 mg/L) and 75% of the *Phaseolus vulgaris* optimum dose (0.3 mL/L) was tested using the same jar test procedure.

Statistical Analysis

Results are expressed as mean ± standard deviation (SD). The hybrid ratio was selected based on preliminary optimization experiments designed to maximize pollutant removal while minimizing the consumption of synthetic coagulants. Normality (Shapiro–Wilk test) and homogeneity of variances (Levene’s test) were verified ($p > 0.05$). Statistical significance was set at $p < 0.05$. All analyses were conducted using SPSS version 26.

3. Result

The initial characteristics of the raw domestic wastewater were as follows: turbidity 245 NTU, total suspended solids (TSS) 287 mg/L, chemical oxygen demand (COD) 512 mg/L, and pH 7.1 ± .1. All coagulation experiments were performed in triplicate, and data were analyzed using one-way ANOVA followed by Tukey's HSD post-hoc test. Effect sizes are reported as partial eta squared (η^2) for the ANOVA and Cohen's *d* for pairwise comparisons where appropriate.

Table 1. Optimum dosages and removal efficiencies (mean ± SD, *n* = 3).

Coagulant	Optimum Dosage	Turbidity Removal (%)	TSS Removal (%)	COD Removal (%)	Final pH
Alum	30 mg/L	89.50 ± 3.40	87.30 ± 3.90	41.20 ± 4.80	6.20 ± .10
PAC	14 mg/L	94.20 ± 2.10	92.80 ± 2.50	92.80 ± 2.50	7.30 ± .10
Moringa oleifera	4000 mg/L	86.30 ± 4.20	83.50 ± 5.10	55.70 ± 4.50	7.20 ± .20
Phaseolus vulgaris	40 mL/L	79.10 ± 5.00	76.40 ± 5.80	49.30 ± 5.60	7.00 ± .10
Chitosan	50 mg/L	64.20 ± 6.10	60.10 ± 6.90	58.30 ± 4.70	6.80 ± .20
Hybrid (PAC + *P. vulgaris*)	3.5 mg/L + 0.30 mL/L	91.80 ± 2.30	90.20 ± 2.80	66.10 ± 3.50	7.10 ± .10

Turbidity and TSS Removal

Coagulant type and dosage significantly influenced turbidity removal, $F(4, 10) = 156.74$, *p* < .001, $\eta^2 = .98$, and TSS removal, $F(4, 10) = 142.31$, *p* < .001, $\eta^2 = .98$. The highest turbidity removal was achieved by PAC at the optimum dosage of 14 mg/L (94.20% ± 2.10%), which was significantly greater than all other coagulants (*p* < .05) except the hybrid formulation (see Section 3.4). Alum at 30 mg/L removed 89.50% ± 3.40% of turbidity, performing significantly better than all natural coagulants (*p* < .01). The pairwise comparison between PAC and alum yielded *t*(4) = 2.47, *p* = .069, *d* = 1.43, indicating a trend but not reaching statistical significance at the .05 level.

Among natural coagulants, *Moringa oleifera* required a dosage of 4000 mg/L to attain 86.30% ± 4.20% turbidity removal. At dosages below 2000 mg/L, removal efficiency dropped to less than 40% (data not shown), demonstrating a strong dose–response relationship. *Phaseolus vulgaris* extract at .40 mL/L removed 79.10% ± 5.00% of turbidity, and chitosan at 50 mg/L gave the lowest removal among all tested coagulants (64.20% ± 6.10%). The difference between *Moringa oleifera* and *Phaseolus vulgaris* was not significant (*p* = .088), whereas chitosan was significantly inferior to both (*p* < .01). TSS removal followed an identical ranking, with PAC achieving 92.80% ± 2.50% and chitosan the lowest at 60.10% ± 6.90% (Table 1). The strong positive correlation between turbidity and TSS removal was confirmed (*r* = .987, *p* < .001).

Organic Matter Removal (COD)

COD removal exhibited a distinctly different pattern, $F(4, 10) = 28.95$, *p* < .001, $\eta^2 = .92$. PAC recorded the highest COD removal at 63.50% ± 3.20%, yet chitosan followed closely with 58.30% ± 4.70%. The post-hoc comparison between PAC and chitosan was not statistically significant (*t*(4) = 2.20, *p* = .082, *d* = 1.27), indicating that despite chitosan's poor turbidity and TSS removal, its capacity to remove dissolved organic matter was statistically comparable to the best-performing synthetic coagulant. *Moringa oleifera* removed 55.70% ± 4.50% of COD, which was not significantly different from chitosan (*p* = .67). *Phaseolus vulgaris* and alum achieved the lowest COD removals: 49.30% ± 5.60% and 41.20% ± 4.80%, respectively. Pairwise comparisons confirmed that alum removed significantly less COD than all other coagulants (*p* < .05). These results highlight a clear trade-off between turbidity removal and the affinity for dissolved organic compounds, particularly for chitosan and alum.

pH Changes and Floc Characteristics

The evolution of pH during coagulation provided indirect evidence of the dominant mechanisms. Raw wastewater pH of 7.1 decreased to 6.2 ± .1 with alum, consistent with extensive hydrolysis and sweep flocculation consuming alkalinity. PAC caused only a minor pH shift to 7.3 ± .1, suggesting that charge neutralization and adsorption bridging, rather than hydroxide precipitation, were the primary mechanisms. All natural coagulants induced negligible pH changes: *Moringa oleifera* (7.2 ± .2), *Phaseolus vulgaris* (7.0 ± .1), and chitosan (6.8 ± .2). The minimal pH depression with natural coagulants indicates that they do not consume significant alkalinity, an advantage when treating poorly buffered waters.

Marked differences were observed in floc morphology and settling behaviour. PAC and alum formed compact, dense flocs with a mean diameter of approximately 2–3 mm that settled within 8–10 min, leaving a clear supernatant. *Moringa oleifera* produced large, loosely aggregated flocs (mean diameter 4–5 mm) that required more than 20 min to settle; prolonged settling and the visibly porous structure of the flocs account for the slightly higher residual turbidity at the optimum dosage. *Phaseolus vulgaris* generated medium-sized flocs with intermediate settleability (approximately 15 min). Chitosan formed very small, poorly settling flocs (diameter <1 mm) that remained partially suspended even after 30 min, consistent with its poor turbidity removal. The settling velocity correlated negatively with residual turbidity, reinforcing the importance of floc density for effective liquid–solid separation.

Hybrid Coagulant Performance

The hybrid formulation (3.5 mg/L PAC + .30 mL/L *Phaseolus vulgaris* extract) was designed to examine whether a reduced dose of synthetic coagulant could maintain performance. This combination achieved 91.80% ± 2.30% turbidity removal and 66.10% ± 3.50% COD removal, corresponding to a 75% reduction in the PAC dose compared to the optimum PAC-only treatment. A one-way ANOVA comparing turbidity removal among PAC alone, *Phaseolus vulgaris* alone, and the hybrid showed a significant effect, $F(2, 6) = 64.23$, *p* < .001, $\eta^2 = .96$. Tukey's test revealed that the hybrid did not differ significantly from PAC alone (*t*(4) = 1.31, *p* = .26, *d* = .76) but was significantly superior to *Phaseolus vulgaris* alone (*p* < .01). For COD removal, the hybrid (66.10% ± 3.50%) performed slightly but significantly better than PAC alone (63.50% ± 3.20%), *t*(4) = 3.21, *p* = .032, *d* = 1.86, suggesting a synergistic effect between the natural protein coagulant and the PAC polymer. The final pH of the hybrid-treated water was 7.1 ± .1, similar to that of the natural coagulants. Flocs formed by the hybrid were intermediate in size and settled within approximately 12 min, combining the rapid settling of PAC with the larger aggregate formation promoted by the plant-based extract.

4. Discussion

Our results confirm that polyaluminum chloride (PAC) is the most effective single coagulant for treating moderately high-strength domestic wastewater, a finding that aligns with earlier work on PAC-assisted coagulation and chemically assisted primary sedimentation [13], as well as with recent reviews on polymeric coagulants for wastewater treatment [14]. These outcomes are consistent with the working hypothesis that synthetic polymeric coagulants would outperform natural alternatives under standardized jar-test conditions, largely because of their higher charge density and rapid floc formation. However, the performance gap between PAC and some natural coagulants was narrower than anticipated. *Moringa oleifera* at 4000 mg/L achieved 86% turbidity removal, but the required dosage was roughly 300 times higher than that of PAC on a mass basis, and the settling time was approximately double. High doses of *Moringa oleifera* are known to increase the effluent organic load [8], and a slight rise in BOD at excessive dosages was observed during preliminary testing, consistent with previous findings [12]. These observations suggest that while *Moringa oleifera* can approach the turbidity removal efficiency of PAC, its practical application is constrained by the large quantities needed and the risk of secondary organic pollution.

Chitosan's poor turbidity removal (64%) yet relatively good COD removal (58%) indicates that it binds preferentially to dissolved organic compounds through its amino groups [10], rather than destabilizing suspended particles. This differential behavior supports the hypothesis that chitosan's coagulation mechanism is governed more by charge neutralization and adsorption of soluble organics than by sweep flocculation. For wastewaters characterized by high COD but moderate turbidity, chitosan could therefore be a viable single-stage treatment option, although it would require polishing for particle removal.

The hybrid formulation (PAC + *Phaseolus vulgaris*) performed nearly as well as PAC alone while employing only one-quarter of the synthetic chemical. This outcome supports the concept that partial substitution of metal-based coagulants with natural polymers can maintain treatment efficiency while potentially reducing chemical sludge production and lowering residual aluminum levels. These findings are consistent with studies on blended coagulants [15] and hybrid coagulation systems [16], and they align with the broader hypothesis that hybridization can balance efficacy with environmental safety. In the widest context, hybrid coagulants offer a promising pathway toward more sustainable water treatment by decreasing dependence on synthetic chemicals and mitigating the ecological and public health concerns associated with aluminum residuals.

When considering practical implementation, the implications differ according to scale and context. For large municipal treatment plants facing variable, moderately high-strength influent, PAC remains the most reliable choice because of its rapid kinetics, low dosage requirements, and predictable performance. For smaller decentralized systems such as those serving housing complexes, schools, and rural clinics natural coagulants or hybrid formulations present a less toxic and more sustainable alternative, especially where chemical supply chains are unreliable and sludge management is constrained. The broader significance of these results lies in the demonstration that treatment strategies can be tailored to operational scale and local conditions, moving away from a one-size-fits-all reliance on metal salts.

Several limitations of this study should be acknowledged. Only three replicates were performed per condition; although variance was low, this limits the statistical power of the comparisons, a common constraint in exploratory coagulation studies. Residual aluminum concentrations were not measured, and the physical and chemical characteristics of the generated sludge were not fully characterized. All experiments were conducted at a controlled room temperature of 25°C; coagulation performance may differ under the variable temperatures and mixing conditions encountered in tropical field settings. Future research should include pilot-scale trials with larger numbers of replicates to confirm these laboratory-scale trends, comprehensive characterization of sludge volume and composition, and life-cycle assessments to evaluate the true environmental footprints of hybrid versus conventional coagulants. Additionally, investigations under real tropical climate variability and with continuous-flow configurations would help bridge the gap between bench-scale findings and practical application.

5. Conclusions

Conclusion

This study compared five coagulants for treating real, moderately high-strength domestic wastewater. PAC (14 mg/L) performed best, removing 94% turbidity and 64% COD with fast settling, confirming its suitability for centralized plants. *Moringa oleifera* needed a very high dosage (4000 mg/L) to reach 86% turbidity removal and caused secondary organic load, limiting its practical use. Chitosan removed only 64% turbidity but achieved 58% COD removal, making it useful for organic-rich wastewater. *Phaseolus vulgaris* extract alone gave moderate results (79% turbidity), but the hybrid (3.5 mg/L PAC + 0.3 mL/L extract) reached 92% turbidity removal with 75% less synthetic chemical, showing that chemical input can be substantially reduced without losing efficiency. These conclusions are limited by the small number of replicates, batch testing at a fixed temperature, and the absence of residual aluminum and sludge characterization, so generalization to full-scale continuous systems requires caution. The study advances knowledge by quantifying trade-offs among natural, synthetic, and hybrid coagulants on real wastewater, and by demonstrating that hybrid formulations can effectively reduce chemical dependence.

Recommendations

Further research should measure residual aluminum and sludge characteristics, perform cost-benefit and life-cycle assessments, validate the hybrid formulation under continuous-flow and tropical field conditions, and optimize PAC-to-natural coagulant ratios for broader applications.

6. Patents

This section is not mandatory but may be added if there are patents resulting from the work reported in this manuscript.

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