Low-Cost Innovative Water Treatment Using Binary Bio-Coagulants: A Sustainable Approach for Turbidity and Hardness Removal in Synthetic Turbid Water

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Abstract - *This study investigates a novel, low-cost approach for treating synthetic turbid water using binary blends of plant-based bio-coagulants, through a comparative assessment against conventional alum. Four plant-derived coagulants were blended in seed ratios of 25:75, 40:60, 50:50, 60:40, and 75:25, and applied at dosages ranging from 50 to 85 mg/L. These blends were tested on synthetic turbid water samples with turbidity levels of 20, 40, 50, and 80 NTU using standard jar test procedures. The study assessed variations in water quality parameters, including turbidity, pH, chloride, and total hardness. Results showed that specific binary combinations achieved turbidity removal efficiencies of up to 90%, matching or exceeding alum performance. Additionally, bio-coagulant treatments maintained pH within the acceptable range of 6.5–8.5, whereas alum led to significant pH reductions necessitating chemical correction. Select combinations also showed notable hardness reduction, offering advantages over conventional coagulants. Unlike alum-based sludge, the sludge generated from bio-coagulants was biodegradable and potentially reusable as organic manure, reducing environmental impact. Among all combinations tested, the Phyllanthus emblica / Cicer arietinum blend exhibited the highest turbidity removal efficiency (up to 90%) while maintaining optimal pH and hardness levels. These findings indicate that binary bio-coagulants, when applied at suitable ratios and dosages, provide a sustainable, cost-effective, and efficient alternative to chemical coagulants, especially suitable for decentralised water treatment applications.*

Keywords: Alum,Jar test,; Turbidity, Hardness, Seed ratio, Binary bio-coagulants.

1. INTRODUCTION

Access to safe drinking water is a critical public health requirement. Turbidity and hardness are among the major concerns in surface and groundwater sources, especially in developing regions. Turbidity, caused by suspended solids such as clay, silt, and organic matter, reduces water clarity and hampers disinfection efficiency. Hardness, primarily due to calcium and magnesium ions, leads to scaling in pipelines and appliances, reduces soap efficiency, and impairs taste [1].

Conventional treatment methods rely on chemical coagulants like aluminium sulphate (alum) to remove turbidity through coagulation–flocculation processes. While effective, alum has several limitations: it significantly lowers water pH, contributes to increased residual aluminium content, and generates large volumes of non-biodegradable sludge that complicate disposal [2-3]. Post-treatment pH correction using lime is often necessary, which raises operational costs and chemical load. Furthermore, concerns regarding long-term health effects of aluminium exposure have driven the search for safer, sustainable alternatives [4].

In recent years, plant-based bio-coagulants have emerged as promising eco-friendly substitutes. Derived from seeds, peels, or pods of various plants, bio-coagulants are biodegradable, non-toxic, locally available, and cost-effective [5-6]. They minimize alteration in pH, produce less sludge, and are especially suitable for decentralized water treatment systems. Various studies have reported the successful application of plant-based bio-coagulants such as *Moringa oleifera*, banana peel powder, and okra mucilage in reducing turbidity and other water contaminants [7]. Recent studies have also demonstrated the effectiveness of plant-based bio-coagulant extracts in treating surface water with comparable performance to conventional coagulants [8].

Despite growing interest, most previous studies have focused on individual plant-based coagulants. However, recent research suggests that binary or hybrid combinations of coagulants can exhibit synergistic effects, potentially improving coagulation performance, reducing required dosage, and broadening the pH range of effectiveness [9-4]. Of particular interest is *Phyllanthus emblica* (Indian gooseberry), which not only aids in turbidity removal but has also demonstrated hardness reduction capability due to its calcium-binding polyphenols [10-11].

To address this gap, the present study evaluates the effectiveness of binary plant-based bio-coagulant blends—

specifically combinations of *Cicer arietinum*, *Trigonella foenum-graecum*, *Dolichos lablab*, and *Phyllanthus emblica*—in treating synthetic turbid water. The study focuses on turbidity removal efficiency, optimal dosage, pH sensitivity, chloride concentration, and hardness reduction, with a comparative analysis against conventional alum. The findings aim to support the development of scalable, sustainable water treatment alternatives suitable for resource-limited settings.

2. MATERIALS AND METHODS

2.1 Jar Test Apparatus

A standard jar test apparatus with six beakers and adjustable stirring speeds was used to evaluate coagulation performance. The setup simulated conventional water treatment steps: rapid mixing for coagulant dispersion, slow mixing for floc formation, and sedimentation for particle settling.

2.2 Preparation of Synthetic Turbid Water

Synthetic turbid water was prepared by dispersing 0.03 g/L of bentonite clay into tap water and mixing vigorously using a magnetic stirrer to ensure uniform dispersion. This preparation method typically yields approximately 1 NTU of turbidity per 0.03 g/L of bentonite under controlled laboratory conditions [12]. By varying the concentration, initial turbidity levels of 20, 40, 60, and 80 NTU were generated—representing field conditions such as storm water runoff and river pollution. Turbidity was measured using a calibrated nephelometric turbidimeter, following standard procedures outlined in APHA [13]. Synthetic samples were allowed to stabilise prior to testing to avoid sedimentation errors.

Note: Although specific targets of 20, 40, 60, and 80 NTU were intended, minor variations were observed due to limitations in synthetic sample preparation. These deviations were minimal and did not significantly affect the comparative performance analysis.

2.3 Preparation, Extraction, and Dosage Determination of Bio-Coagulants

Seeds of *Cicer arietinum*, *Dolichos lablab*, *Trigonella foenum-graecum*, and *Phyllanthus emblica* were sourced locally, thoroughly cleaned, shade-dried, and ground into a fine powder using a laboratory grinder. For extract preparation, 10 g of seed powder was mixed with 100 mL of distilled water and stirred using a magnetic stirrer for 30 minutes. The resulting suspension was filtered through a muslin cloth (pore size ~100–150 µm), following APHA protocols [13], to remove coarse particles. The filtrate was used immediately in coagulation tests.

Binary plant-based bio-coagulant blends were prepared in five specific weight ratios: 25:75, 40:60, 50:50, 60:40, and 75:25 (w/w). To study the effect of varying seed proportions, these blends were tested at dosages of 50, 70, 75, 80, and 85 mg/L. Each dosage was selected to correspond with one of the target initial turbidity levels: 20, 40, 60, and 80 NTU respectively. For each turbidity level, six beakers were used—five containing the binary coagulant blends and one control without any coagulant to assess natural sedimentation.

The dosage range was informed by prior research, which indicates that plant-based coagulants typically achieve optimal turbidity removal within 25–100 mg/L, depending on the source material and water characteristics [12-6-9]. Hence, the dosages used in this study fall within scientifically validated limits and were further refined through preliminary testing.

2.4 Experimental Procedure

Each experiment was carried out using six 1 L beakers of synthetic turbid water placed in individual beakers. The standard coagulation–flocculation process involved the following steps:

- Rapid mixing at 150 rpm for 3 minutes (coagulant dispersion)
- Slow mixing at 30 rpm for 30 minutes (flocculation)
- Settling for 45–60 minutes (sedimentation phase)

Following sedimentation, the supernatant was analysed for turbidity, pH, chloride concentration, and total hardness.

2.5 Analytical Methods

All water quality parameters were measured in accordance with standard methods:

- Turbidity: Nephelometric method
- pH: Digital pH meter
- Chlorides: Mohr's titration method
- Hardness: EDTA titration (expressed as mg/L $CaCO_3$)

All tests were conducted in triplicate to ensure reproducibility. The average values from each set were used for final analysis.

Note: Suspended solids were not measured as coagulation is typically followed by filtration, which removes residual particulates.

3. RESULTS AND DISCUSSION

This section presents the comparative performance of binary bio-coagulant combinations and conventional alum in treating synthetic turbid water. The parameters evaluated include turbidity removal efficiency, post-treatment pH, changes in chloride concentration, and total hardness. Each combination was tested across a range of seed ratios and dosages under controlled laboratory conditions using the standard jar test method. The results are discussed with reference to graphical data and supported by relevant literature. The aim is to evaluate the technical feasibility, environmental benefits, and operational advantages of binary bio-coagulants as sustainable alternatives to conventional chemical coagulants in water treatment applications.

3.1 Turbidity Removal Efficiency

Turbidity removal is a primary parameter for evaluating coagulant performance. Four binary bio-coagulant combinations and alum were tested across varying initial turbidity levels (20–80 NTU). As shown in Figure 1, the *Phyllanthus emblica / Cicer arietinum* blend achieved the highest average turbidity removal (90%), followed by *Cicer arietinum / Trigonella foenum-graecum* (89.3%) and *Dolichos lablab / Phyllanthus emblica* (87.9%). These values are comparable to, or slightly better than alum (89.4%) under identical conditions.

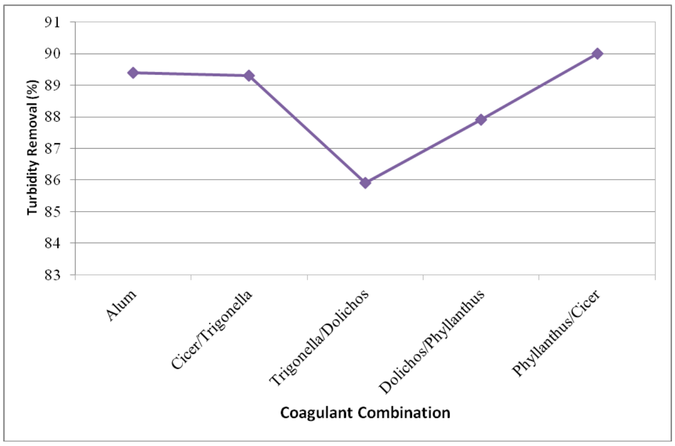

Figure 1. Turbidity Removal Efficiency (%) of Alum and Binary Bio-Coagulant Combinations.

Among the tested combinations, *Trigonella foenum-graecum / Dolichos lablab* yielded the lowest turbidity removal (85.9%), which was notably below alum (89.4%). This relatively reduced efficiency may be attributed to a lower concentration of effective coagulating compounds—such as polyphenols and cationic proteins—in this blend. *Trigonella* seeds, although known for saponins, exhibit lesser flocculating potential compared to the tannin-rich *Phyllanthus emblica*. Similarly, *Dolichos lablab* alone does not significantly contribute to bridging or sweep flocculation without a strongly active coagulant partner. The observed dip highlights that not all binary combinations yield synergistic results. Hence, rational pairing of seed types based on their phytochemical profiles is essential to maximise coagulation efficiency. The lower turbidity removal by *Trigonella foenum-graecum / Dolichos lablab* (85.9%) is in line with prior studies [14-15], which report limited coagulation potential of these seeds individually. As noted by Choy et al. [12], binary combinations without synergistic biochemical interaction may fail to surpass conventional coagulants like alum.

The trend in Figure 1 demonstrates that combinations containing *Phyllanthus emblica* consistently achieved higher removal across all tested turbidity levels. This highlights the enhanced performance due to the presence of polyphenols, tannins, and proteins. The *Phyllanthus emblica / Cicer arietinum* blend, in particular, exhibited superior turbidity removal, likely due to polyphenol-mediated bridging from *Phyllanthus emblica* and protein-based flocculation contributed by *Cicer arietinum*. Similar observations were reported by Chowdhury et al. [7], who demonstrated that combinations of agricultural bio-coagulants provided improved turbidity reduction. Nur et al. [9] further observed that binary coagulants with complementary bioactive compounds enhanced floc formation. Yin et al. [16] confirmed that multifunctional plant-based polymers could match or exceed the performance of alum when properly dosed.

Unlike alum, which typically demands post-treatment pH correction, all bio-coagulant blends maintained high turbidity removal without chemical adjustment, making them suitable for decentralised, low-cost water treatment systems.

3.2 Post-Treatment pH

Maintaining an acceptable pH range in treated water is critical, as deviations can affect taste, increase corrosion potential, and impair disinfection efficiency. According to WHO [1], the recommended pH range for drinking water lies between 6.5 and 8.5. In this study, all coagulant treatments were assessed for their impact on post-treatment pH across different turbidity levels.

As illustrated in Figure 2, conventional alum consistently reduced the pH of treated water to a range between 6.4 and 6.6, especially at higher dosages. This trend aligns with Ahmad et al. [2], who reported that alum hydrolyses to form acidic species. Specifically, aluminium ions react with water to produce aluminium hydroxide and release hydrogen ions, which in turn lower the pH. Such acidic conditions often necessitate post-treatment lime addition to restore pH to acceptable levels, increasing chemical usage and operational costs [3-4].

In contrast, all binary bio-coagulant blends maintained post-treatment pH values within the safe range of 6.5 to 8.5. Notably, the *Phyllanthus emblica / Cicer arietinum* blend exhibited a slightly alkaline trend, with pH values ranging from 7.62 to 8.18 across all tested turbidity levels

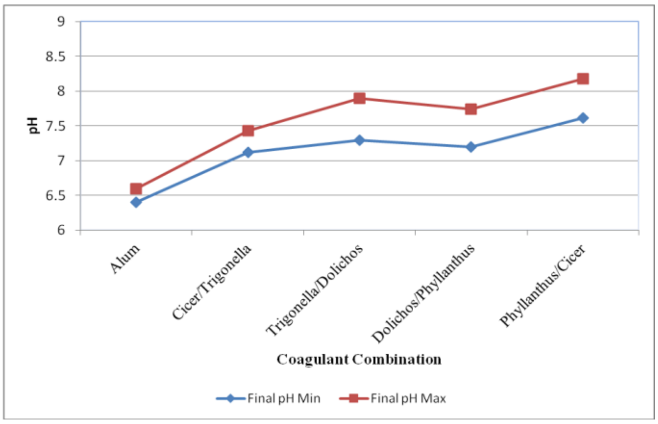

Figure 2. Post-Treatment pH of Treated Water Using Different Coagulants.

This stabilising effect is attributed to the presence of organic acids and polyphenolic compounds in *Phyllanthus emblica*, which impart mild alkalinity and buffering capacity [10-11]. These findings are supported by Radoiu et al. [17], who observed that plant-based coagulants typically contain tannins, proteins, and natural buffers that help minimise pH fluctuations during coagulation. Similarly, Yin et al. [16] noted the inherent pH stability of natural coagulants, making them particularly suitable for decentralised systems where chemical correction is not feasible or cost-effective.

Furthermore, bio-coagulants promote downstream disinfection efficiency by maintaining near-neutral pH conditions. Chlorination, a widely used disinfection method, is most effective within a pH range of 6.5 to 7.5, where hypochlorous acid formation is optimal. Since alum frequently reduces pH below this effective window, additional alkaline dosing is required to enable effective disinfection. By contrast, the natural buffering action of plant-based coagulants simplifies post-treatment requirements and enhances process sustainability [6-17-18].

3.3 Post Treatment Chloride Level

Monitoring chloride concentrations after treatment is crucial, as elevated levels can lead to corrosivity in pipelines, poor taste, and potential non-compliance with drinking water standards. The World Health Organization (WHO) prescribes a maximum allowable chloride concentration of 250 mg/L in potable water [1]. Accordingly, this study analysed the chloride levels in water treated with both binary bio-coagulants and alum to determine whether the coagulants introduced additional salinity.

As illustrated in Figure 3, all treated samples remained well within the WHO limit, including those processed with the *Phyllanthus emblica / Cicer arietinum* blend. Although a minor increase in chloride content was observed across all samples, the maximum recorded value was 20.5 mg/L—associated with the *Dolichos lablab / Phyllanthus emblica* blend at a dosage of 85 mg/L. These slight elevations are attributed to the presence of naturally occurring chloride salts and organic acids in plant-based coagulant materials, particularly in *Phyllanthus emblica* [19].

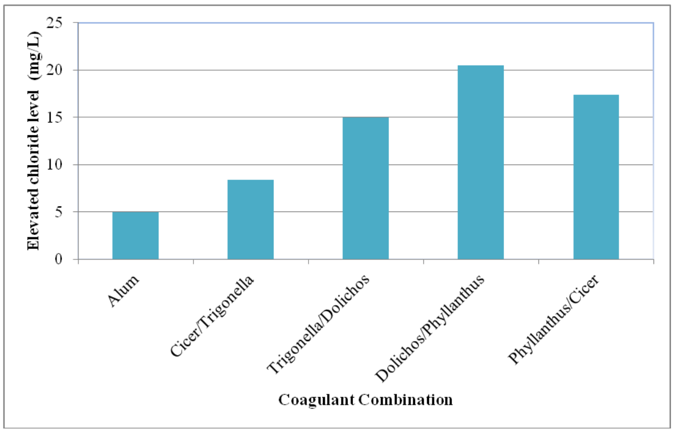

Figure 3. Increase in chloride level after Coagulation with Alum and Plant-Based Coagulants.

Importantly, none of the treatments approached the regulatory threshold, and the changes observed were less pronounced compared to those caused by alum, especially at higher dosages. This observation supports the findings of Okuda et al. [20] and Choy et al. [12], who reported that plant-based coagulants—especially those derived using saline extraction—may lead to minimal chloride increases but remain safe for consumption. In contrast, alum hydrolysis can elevate ionic strength in the water, which may enhance chloride reactivity and contribute to long-term corrosion in metallic distribution systems [17]. The comparatively lower chloride elevation observed with bio-coagulants underscores their suitability for long-term use in drinking water systems, particularly in decentralised or rural regions. In areas such as Tindivanam, Tamil Nadu, where surface and groundwater chloride levels are already modest (20–80 mg/L), even minimal additions remain safely within permissible limits. However, total hardness often exceeds national standards, posing greater water quality challenges [21-22].

This regional hydro-geochemical profile further enhances the relevance of binary bio-coagulants that address both turbidity and hardness without exacerbating chloride content. Combinations such as *Dolichos lablab / Phyllanthus emblica* and *Phyllanthus emblica / Cicer arietinum* demonstrated dual efficacy and are especially suitable for areas with high hardness but low chloride levels.

3.4 Hardness Reduction

Hardness in water, primarily due to calcium and magnesium ions, poses a significant challenge in regions such as Tindivanam, where surface and groundwater sources often exhibit hardness levels exceeding 300 mg/L [23-24]. Excessive hardness contributes to pipe scaling, reduced soap efficiency, undesirable taste, and long-term operational issues in both domestic and industrial settings.

As shown in Figure 4, the binary blend of *Dolichos lablab / Phyllanthus emblica* achieved the highest total hardness reduction, with a decrease of 59.8 mg/L. The Phyllanthus emblica / Cicer arietinum combination followed with a reduction of 27.5 mg/L. By contrast, alum-treated samples exhibited negligible hardness removal, and in some cases, even a slight increase—likely due to ionic exchanges or the presence of residual aluminium compounds.

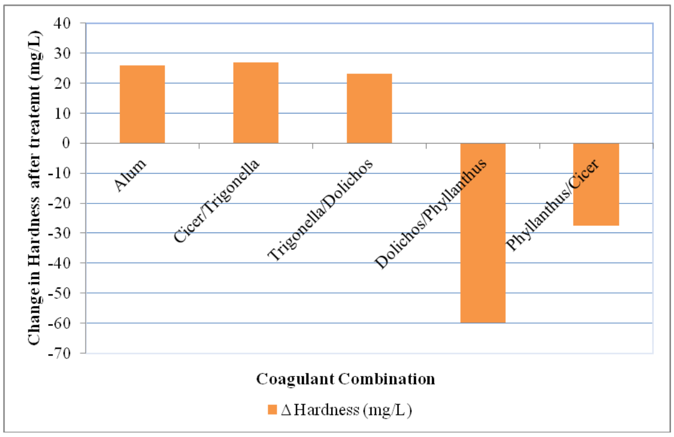

Figure 4. Change in Hardness after Coagulation Indicating Softening Effect of *Phyllanthus*-Based Blends.

The effective hardness reduction observed in *Phyllanthus emblica*-based blends can be attributed to its content of polyphenols, citric acid derivatives, and natural chelators, which have calcium- and magnesium-binding affinity [25-10]. These compounds likely contribute to partial water softening by destabilizing divalent cations during floc formation. Similarly, the high protein and polysaccharide content of *Dolichos lablab* aids in flocculation and entrapment of hardness ions [26].

Recent studies have validated the dual functionality of bio-coagulants—turbidity removal and hardness reduction—when applied at optimised dosages. This supports their relevance in settings such as Tindivanam, where water often displays persistent hardness but comparatively low chloride levels [27-24]. Unlike alum, which lacks any softening effect and may increase the ionic load, bio-coagulants offer a sustainable, multi-functional alternative. Their inherent buffering and chelating capacity also improves water quality without elevating total dissolved solids (TDS) or leaving behind harmful residuals [17-26].

3.5 Sludge Characteristics

The characteristics of sludge generated during coagulation–flocculation are critical in evaluating both the environmental impact and operational feasibility of water treatment processes. Alum-based treatments are known to produce inorganic, non-biodegradable sludge that necessitates careful disposal due to the presence of residual aluminium and associated health risks [2].

In contrast, the sludge generated from plant-based bio-coagulants is organic in nature, biodegradable, and environmentally benign. In the present study, sludge volumes produced by binary bio-coagulants were significantly lower and settled more easily compared to those from alum treatment. Notably, flocs formed using *Dolichos lablab* and *Phyllanthus emblica* were more compact and denser, facilitating faster sedimentation and reduced bulk.

These observations are consistent with the findings of Yin et al. [16], Kumar et al. [19], and Radoiu et al. [17], who reported that plant-based coagulants yield less voluminous, stable flocs that settle more rapidly due to natural polymers and mucilaginous compounds. Furthermore, the organic nature of bio-coagulant sludge opens avenues for reuse in agriculture as a soil conditioner or compost additive, particularly when the sludge originates from potable water treatment processes.

Studies involving sludge derived from *Moringa oleifera* and similar plant extracts have shown that such materials can be safely applied to soil without causing toxicity, while also enhancing organic content and water retention capacity [7-26]. In contrast, alum-based sludge is chemically reactive, requiring secure drying and regulated disposal. Its accumulation increases operational costs, particularly in decentralised and rural water supply systems.

Thus, the use of biodegradable coagulants contributes not only to waste minimisation but also aligns with circular economy principles by transforming a treatment by-product into a beneficial agricultural input. In line with circular economy principles, several studies have explored recovery and reuse of sludge-based coagulants for sustainable water treatment applications [28]. Shawal et al. [29] conducted parametric studies on coagulant recovery from water treatment sludge for circular economy integration. Additionally, valuable elements in sludge have been evaluated for recovery potential in municipal wastewater systems [30].

3.6 Summary of Performance of Binary Bio-Coagulant Combinations

Figure 5 presents a consolidated graphical comparison of the performance of all four binary bio-coagulant blends and alum across five critical water quality parameters: turbidity removal efficiency, post-treatment pH, chloride concentration, total hardness reduction, and sludge characteristics.

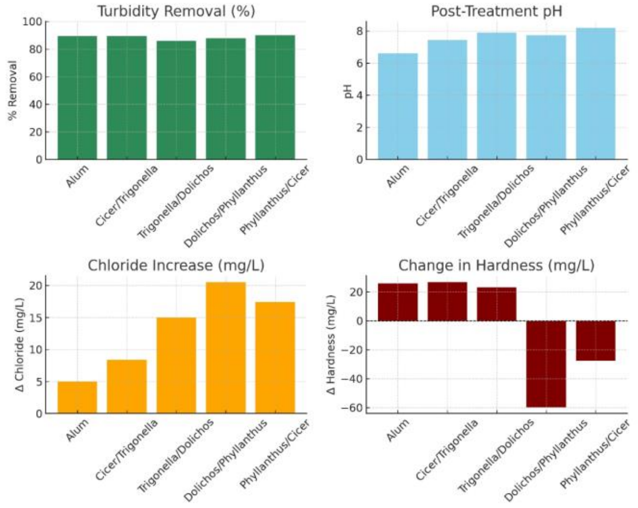

Figure 5. Comparative Performance of Coagulants across Key Water Quality Parameters.

Among all blends, the *Phyllanthus emblica / Cicer arietinum* combination achieved the highest turbidity removal (90%), as shown in Figure 5 (top-left). This is marginally better than alum (89.4%) and the *Cicer / Trigonella* blend (89.3%). The lowest turbidity removal was recorded for *Trigonella / Dolichos* (85.9%), indicating that not all combinations yield synergistic benefits. The variation highlights that combinations involving *Phyllanthus emblica* consistently outperform others. This reinforces its dominant role in floc formation, likely due to its high polyphenol and organic acid content, which enhance charge neutralisation and particle bridging. The blends also maintained removal efficiency across all tested turbidity levels, whereas alum performance fluctuated more with initial turbidity. Thus, turbidity removal trends, as reflected in Figure 5, reaffirm the suitability of certain binary blends as robust alternatives to conventional alum.

All binary blends consistently maintained post-treatment pH within the WHO-recommended range (6.5–8.5), while alum consistently reduced pH to between 6.4 and 6.6. This aligns with earlier findings confirming the natural pH-stabilising behaviour of plant-based coagulants [16-4]. Regarding chloride levels, all blends exhibited minimal increases, with the highest recorded value being 20.5 mg/L—well below the 250 mg/L WHO threshold. These increases are attributed to trace chloride content in the seed matrix and were substantially lower than those caused by alum [12-20].

In terms of hardness removal, the *Dolichos lablab / Phyllanthus emblica* combination emerged as the most effective, with a reduction of 59.8 mg/L, followed by *Phyllanthus emblica / Cicer arietinum* at 27.5 mg/L. The enhanced performance is likely due to the chelating nature of citric acid derivatives and polyphenols in *Phyllanthus emblica*, coupled with sweep flocculation facilitated by *Dolichos lablab*. By contrast, alum exhibited no appreciable softening and in some instances slightly increased hardness—possibly due to ionic shifts during coagulation [24].

Finally, sludge characteristics observed during experimentation demonstrated clear advantages for all bio-coagulant combinations. The bio-sludge settled faster, was lower in volume, and fully biodegradable, offering opportunities for composting or soil application. Alum-based sludge, in contrast, was bulkier, required careful handling, and lacked reuse potential, as supported by Radoiu et al. [17] and Usman et al. [26]. Overall, the synthesis presented in Figure 5 reinforces the technical and environmental superiority of binary bio-coagulant blends—particularly those involving *Phyllanthus emblica*. These findings not only validate their practical potential for rural and semi-urban water treatment but also position them as promising components in the transition toward green, decentralized water purification systems [26-7].

The growing emphasis on bio-coagulants is further supported by recent reviews. Tijjani Usman et al. [31] discussed the synergistic effects of combining plant-derived extracts with complementary flocculating compounds to enhance turbidity removal and overall water quality. Similarly, Simate and Ndlovu [32] emphasized the importance of biodegradable, non-toxic coagulants in improving long-term sustainability, especially in decentralized treatment systems.

4. CONCLUSION

This study confirms the effectiveness of binary plant-based bio-coagulants—particularly *Phyllanthus emblica* and *Cicer arietinum*—as sustainable, low-cost alternatives to alum for treating synthetic turbid water. Optimised seed ratios of 25:75 and 40:60 achieved up to 90% turbidity removal while maintaining post-treatment pH within the WHO-recommended range of 6.5–8.5, eliminating the need for additional pH correction. Combinations involving *Dolichos lablab* yielded notable hardness reductions up to 59.8 mg/L, highlighting their potential for regions with high water hardness. Chloride levels remained well within permissible limits, and the biodegradable sludge settled rapidly and showed promise for agricultural reuse.

The findings strongly support the application of binary bio-coagulants in decentralised and rural water treatment systems, where affordability and multi-parameter water quality management are crucial. This novel combinatory approach offers synergistic advantages in coagulation efficiency, pH stability, and ion removal—advancing beyond prior studies that focused on individual coagulants.

Future research should include pilot-scale validation, sludge characterisation via SEM imaging, and nutrient analysis to confirm agricultural safety. Moreover, integrating AI-driven predictive modeling could optimise coagulant dosage and blend ratios based on real-time water quality inputs, enabling automation and scalability in smart water treatment systems. Such innovations align with sustainable development goals and circular economy principles, offering environmentally sound solutions for communities with limited resources.

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