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Original Research

Coagulation Potential of Leaf-Based Natural Materials in the Treatment of Industrial Pharmaceutical Wastewater

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ABSTRACT

The growing demand for healthcare and population increase has accelerated the pharmaceutical industry, resulting in large-scale drug production. Consequently, partially or fully treated effluents are often released into the environment, posing risks to both ecosystems and human health. Pharmaceutical wastewater, particularly antibiotics, significantly contaminates aquatic habitats. The present study investigates the use of plant-based natural coagulants (NC) as a low-cost, sustainable, and eco-friendly alternative to synthetic chemicals for the treatment of industrial pharmaceutical wastewater. Among the various natural coagulants, *Moringa oleifera* (moringa leaves) and *Mangifera indica* (mango leaves) were selected. Jar test experiments were performed by mixing a 3 ml stock solution of each coagulant with 1000 ml of pharmaceutical wastewater collected in a beaker. The treatment was conducted under varying hydraulic retention times (HRT) of 2, 4, 6, and 8 hours using coagulant powders of 150 µm size. Parameters such as pH, turbidity, TS, TDS, TSS, COD, alkalinity, phenolic compounds, and phosphate were analyzed. The results indicated that moringa leaf powder achieved the highest COD reduction of 87.6% at 8 hours HRT. Furthermore, sludge obtained after treatment with moringa at 8 hours HRT was analyzed using FE-SEM and EDX to evaluate structural morphology and elemental composition.

INTRODUCTION

The pharmaceutical industry in India has perceived evolution in recent times. This progress is mostly determined by the country's swiftly escalating inhabitants and the growing demand for healthcare products and amenities. As a result, the manufacture of pharmaceutical products has also enlarged significantly to meet these mounting needs (Dafale et al. n.d.). Though, the intensification in pharmaceutical manufacturing has led to a equivalent increase in the production of industrial effluents. Through the manufacture course, huge volumes of effluent are liquidated, encompassing a variety of chemical and pharmaceutical deposits. This wastewater is one of the foremost contributors to industrial water pollution in India. According to reports, pharmaceutical wastewater accounts for approximately one-third of all water pollution connected with industrial discharges in the country (Maurya and Daverey 2018). The occurrence of injurious pharmaceutical compounds in the wastewater stances a stern threat to the quality of natural water bodies such as rivers, lakes, and groundwater sources. Additionally, wastewater disposed of from pharmaceutical industries regularly encompasses constituents like antibiotics and organic pollutants. Regrettably, conservative treatment methods do not always succeed in eliminating all antibiotic residues entirely from the wastewater. The ability of pollutant deletion often differs based on the treatment process employed and the scale of the pharmaceutical industry. Recent studies have explored sustainable and low-cost adsorbents for the removal of heavy metals from water systems. For example, Verma and Sachdeva (2025) investigated the use of peanut hulls as a biosorbent for lead, demonstrating its potential in water purification applications. Similarly, Rajput, Benjwal, and Pandey (2025) provided a comprehensive review on bioremediation techniques for heavy metal contamination, highlighting the role of biological approaches in mitigating environmental pollution. In some circumstances, wastewater is predisposed of directly into nearby water bodies without suitable treatment. This practice can have severe negative effects on aquatic ecosystems. It also poses serious risks to human health, such as genotoxicity, hormonal disruption, and overall water toxicity (Desta, Heliyon, 2021).To address this issue, alternative treatment methods are being explored. One such promising method is biological action, which uses naturally occurring microorganisms to reduce contaminants in the wastewater. Contrasting conventional chemical treatment procedures, biological methods bid a more environmentally friendly and cost-effective resolution (Singh et al. n.d.). The use of biological treatment methods helps diminish pollution in aquatic atmospheres. It also diminishes the reliance on synthetic chemicals, making it a more sustainable and safer option for treating pharmaceutical wastewater. Thus, the biological tactic not only supports environmental fortification but also associates with the goals of low-cost and effective wastewater administration.

Orthodox treatment approaches have been extensively adopted to remove pharmaceutical impurities from wastewater. Few of the usually applied techniques comprise ozonation, reverse osmosis, activated carbon filtration, and flocculation. Further, more progressive methods such as membrane bioreactors (MBR) and advanced oxidation processes (AOPs) have also been established to augment the efficiency of pharmaceutical waste removal. Despite the availability of these technologies, the complete removal of pharmaceutical drugs from wastewater remains inconsistent. Their efficiency can vary depending on the type of pharmaceutical compound and the treatment conditions. Monitoring the presence and concentration of these compounds in both water treatment and wastewater treatment plants is a major challenge. This difficulty arises because pharmaceuticals entering wastewater treatment systems are often not fully metabolized by the human body. As a result, active pharmaceutical residues remain in the effluent and pass through the treatment process (Kebede, Dube, and Nindi 2018).One of the chemical methods used in wastewater treatment is coagulation, commonly using coagulants like aluminum sulfate (alum). Aluminum sulfate quickly hydrolyzes when added to an aqueous solution. It forms positively charged cationic species that interact with the negatively charged colloidal particles present in wastewater. This interaction promotes the formation of microflocs through a process known as charge neutralization. However, these microflocs are typically very small and fragile. They lack adequate structural integrity and are disposed to defiance apart when endangered to physical stress, such as agitation or flow turbulence. In addition, the use of aluminum-based coagulants habitually results in the occurrence of residual metal ions in the treated water. This can rise the health and environmental alarms, particularly if the metal concentration surpasses permissible limits. tamarind fruit shells (TFS) used in a fixed-bed reactor can remove over 90% of malachite green dye from aqueous solutions under optimized flow rate, biosorbent size, and bed depth conditions, with bed efficiency exceeding 90% and effective regeneration using 0.5 N HCl and H_2SO_4 (Murugesan et al. 2022). Furthermore, the overall effectiveness of the process manages to be comparatively low under certain working conditions. These limits to underscore the importance of selecting suitable coagulants and optimizing the coagulation–flocculation process to improve treatment performance.

As research continues, more sustainable and effective alternatives to traditional chemical coagulants are being explored (Eri, Hadi, and Slamet 2018).

The coagulation process is used to react with organic compounds in raw wastewater and destabilize suspended particles. Finding alternative coagulants from natural sources that are more affordable and environmentally friendly is essential to solving chemical coagulant issues (Kakoi et al. n.d.). Plants based Natural coagulants such as Moringa oleifera and water hyacinth (Ali et al. n.d.; Camacho et al. n.d.; Sanmuga Priya and Selvan 2014) okra, fenugreek (Lanan et al. n.d.) banana (Kakoi et al. n.d.) cassava (Lugo-Arias et al. 2017) Neem leaf (Ahmad et al. 2021) and acorn (Antov et al. 2018; Šćiban et al. 2009). Even activated carbon from avocado seed waste has good sorption capacity of manganese and chromium ions from contaminated water (Mabalane, Shooto, and Thabede 2024). When natural coagulants utilized to treat various wastewaters, have shown good efficacy in eliminating turbidity, TSS, color, and COD. In this work, the pharmaceutical wastewater was treated using Moringa and Mango Leaves as natural coagulants with Raw Industrial Pharmaceutical wastewater.

2. MATERIALS AND METHODS

2.1 Sampling and characterization of wastewater

Untreated pharmaceutical wastewater samples were obtained straight from a pharmaceutical manufacturing facility. The collection occurred prior to the wastewater entering the current treatment plant. This guaranteed that the samples accurately reflected the untreated effluent produced throughout the production process. Sampling occurred on two distinct days to allow for potential fluctuations in effluent properties. The collected samples were designated as Sample 1 and Sample 2, corresponding to their respective collection days. Subsequent to collection, both samples underwent a comprehensive physico-chemical investigation. This investigation sought to ascertain the fundamental attributes of the untreated pharmaceutical wastewater. The examined parameters encompassed pH, turbidity, total solids (TS), total dissolved solids (TDS), total suspended solids (TSS), chemical oxygen demand (COD), alkalinity, phosphate concentration, and the presence of phenolic compounds. The findings of this preliminary characterization are encapsulated and displayed in Table 1. The parameters of pharmaceutical wastewater exhibited considerable variation between Sample 1 and Sample 2. This fluctuation is ascribed to discrepancies in daily production activities, the nature of raw materials utilized, and alterations in operational conditions within the facility. Subsequent to the analysis, each sample was subjected to treatment with a distinct natural coagulant. Sample 1 was subjected to Moringa leaf powder treatment, recognized for its superior coagulation capabilities and environmental sustainability. Conversely, Sample 2 was subjected to Mango leaf powder, an efficacious plant-derived coagulant with potential use in wastewater treatment. The efficacy of each natural coagulant in diminishing contaminants and enhancing water quality was assessed by jar test studies. The treatment outcomes were evaluated to determine the comparative efficacy of Moringa and Mango leaves as natural coagulants in the treatment of pharmaceutical wastewater.

Fig. 1: Photographic views of Raw Pharmaceutical Wastewater collected from the Industry.

Table 1: Initial Physicochemical Characteristics of Pharmaceutical Wastewater

Sl No	Parameters	Pharmaceutical wastewater Sample 1	Pharmaceutical wastewater Sample 2	Units
1	pH	7.65	7.7	-
2	Turbidity	94	163	NTU
3	Total solids	1196	2000	mg/l
4	Suspended solids	262	1392	mg/l
5	Total Dissolved Solids	908	632	mg/l
6	Volatile suspended solids	169	171	mg/l
7	Total alkalinity	342	2499	mg/l
8	Chlorides	167	391	mg/l
9	Dissolved oxygen	3.2	3.7	mg/l
10	COD	5547	5977	mg/l
11	BOD	1100	1271	mg/l
12	Phosphate	18	26	mg/l
13	Phenolic Compound	BLQ(LOQ:0.002)	BLQ(LOQ:0.002)	

* BLQ (LOQ: 0.002) Below the Limit of Quantification

2.2 Natural Coagulants

This study employed powdered natural coagulants for the treatment of raw pharmaceutical effluents. The designated natural coagulants incorporated Moringa leaves **(***Moringa oleifera***)** and Mango leaves **(***Mangifera indica***)**. These plant-based materials were selected owing to their higher adsorbing capacities related to many other natural ingredients. Numerous research studies have specified that the principal mechanisms involved in the coagulation process using natural coagulants are polymer linking and charge nullification (Aziz et al. 2023; Kumar, Othman, and Asharuddin 2017; Nath, Mishra, and Pande 2021). These mechanisms enable the accumulation of suspended particles, leading to enhanced floc formation and sedimentation.

Natural plant-based coagulants are composed of a complex mixture of lipids, proteins, carbohydrates, as well as polymers of polysaccharides and amino acids. These biopolymers contribute significantly to their ability to destabilize particles in wastewater. In particular, polymer adsorption plays a crucial role in enhancing surface interactions, particle dispersion, and overall flocculation performance. Polymer adsorption is typically evaluated by observing the rate at which the concentration of a solution decreases when it comes into contact with a surface (Villaseñor-Basulto et al. 2018). This property is vital for understanding and optimizing the efficiency of coagulants in the water treatment process. For this study, fresh Moringa and Mango leaves were collected from local trees. The collected leaves were first thoroughly washed to remove dust and impurities. They were then dried naturally under direct sunlight for several days until completely dehydrated. Once dried, the leaves were ground into a fine powder using a mechanical grinder. The resulting powder was sieved through a 150 µm mesh to ensure uniform particle size, which is critical for consistency in dosage and performance. To prepare the stock solution, a dosage of 3 grams of the powdered natural coagulant was measured and dissolved in 100 ml of distilled water. The prepared solution was thoroughly mixed to ensure complete dispersion of the particles. This stock solution was then used in the jar test experiments for treating the pharmaceutical wastewater. The preparation process is illustrated in Figure 3**.**

Fig. 2: Photographic view of Dried Leaf and Powdered form of Moringa and Mango Leaf of size 150 µm

Fig. 3: Photographic view of stock solution prepared by using Mango and Moringa leaves

Moringa and Mango leaves are known to contain bioactive proteins that play a crucial role in the coagulation process. These proteins have the ability to neutralize negatively charged particles suspended in the wastewater, which is a key step in the removal of contaminants. When added to pharmaceutical wastewater, the bioactive proteins interact with the suspended particles, reducing their surface charge and making them more likely to come together. This charge neutralization process enhances the aggregation of fine particles into larger clusters. In addition to neutralizing charges, the proteins also act through a bridging mechanism**.** In this mechanism, the long-chain protein molecules connect multiple small particles, forming larger flocs or clusters. These flocs are heavier and more stable, which allows them to settle more easily at the bottom of the treatment container or sedimentation tank (Aziz et al. 2023; Villaseñor-Basulto et al. 2018).Thus, the presence of these naturally occurring bioactive compounds in Moringa and Mango leaves contributes significantly to their effectiveness as natural coagulants in the treatment of pharmaceutical wastewater.

2.3 Coagulation (Jar Test)

To evaluate the coagulation efficiency of natural coagulants, a jar test procedure was conducted using Moringa and Mango leaf powders. Initially, 3 grams of dried and finely powdered leaves, with a particle size of 150 µm, were measured for each type of leaf. The powdered material was then mixed with 100 ml of distilled water to prepare a stock solution of the coagulant. From the prepared stock solution, varying volumes ranging from 1 ml to 6 ml were taken and added to six separate beakers, each containing 1000 ml of raw pharmaceutical wastewater. These six beakers were placed on a jar test apparatus (Flocculator), as shown in Figure 4. The coagulation process began with rapid mixing of all the samples. The flocculator was operated at a high speed at 100-120 rpm for the first 2 to 3 minutes to ensure uniform dispersion of the coagulant in the wastewater. After the initial rapid mixing, the speed was reduced to allow slow mixing of 30-40 rpm for 15 minutes. This step facilitates the gentle collision and aggregation of particles, promoting the formation of flocs. Once the mixing process was completed, the unit was switched off, and the samples were allowed to settle undisturbed for 30 minutes. This settling period enabled the flocs to settle at the bottom of each beaker, leaving clearer water above. Among the various dosages tested, it was observed that the addition of 3 ml of stock solution resulted in the best coagulation efficiency. This dosage provided the most effective settlement of suspended particles within the 30-minute settling time, as compared to the other dosages tested. This optimal dosage was used in subsequent experiments for performance analysis of both Moringa and Mango leaf coagulants.

Fig. 4: Jar Test Apparatus

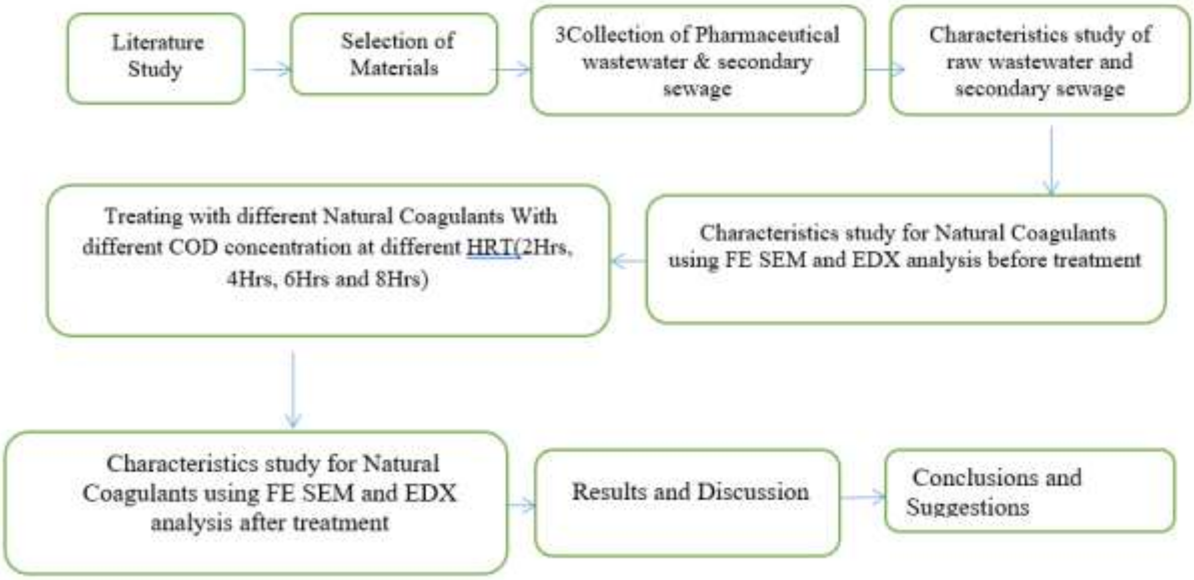
2.4 Comparative Analysis of Moringa (Moringa oleifera) and Mango (Mangifera indica) Leaves as Natural Coagulants

Moringa leaves and Mango leaves, both of which are locally obtained, natural products, have been tested for their efficacy as coagulants for the treatment of pharmaceutical wastewater. The two plant-derived coagulants differ in terms of their chemical composition and mode of coagulation. The Moringa leaves are mainly based on the presence of cationic proteins for coagulation. The proteins are positively charged, and they assist in neutralizing negatively charged suspended particles in wastewater. In contrast, Mango leaves have polyphenols, flavonoids, and tannins that are responsible for their coagulating properties through adsorption and charge neutralization processes. These biochemical content differences affect the effectiveness of the two types of leaves in the coagulation process. Moringa's charge neutralization capacity is greater than that of Mango. This is due to its high cationic protein content, which is more efficient in destabilizing colloidal particles. Conversely, Mango leaves have a moderate charge neutralization because of the concentration and type of their active compounds.

When it comes to turbidity removal, Moringa leaf powder is more efficient. Experimental research has proven that it can remove turbidity by as much as 90 percent. Mango leaf powder, though effective, has an efficiency of turbidity removal between around 50 and 70 percent, which is significantly lower compared to Moringa. Another important consideration is the pH range under which each coagulant functions best. Moringa leaves can operate over a broad pH range, being more suitable for practical application to varying wastewater conditions. Mango leaves are more efficient in near-neutral to slightly acidic conditions, which could be a hindrance for their application in strongly acidic or alkaline wastewater. Floc generation is another point of difference where the two natural coagulants vary. Moringa enhances floc formation more efficiently because of its polymer bridging effect and efficient charge neutralization. This has the effect of producing dense, large, and stable flocs that settle well when undergoing sedimentation. Compared to this, Mango leaves produce smaller and less stable flocs. This is because there is moderate availability of bridging compounds in Mango, and therefore lower flocculation efficiency and lower sedimentation performance.

Notwithstanding these disparities, Moringa and Mango leaves are biodegradable, non-toxic, and eco-friendly. Their natural background renders them environment-friendly alternatives to the standard chemical coagulants, which tend to bring toxicities and sludge-disposal issues.Moringa oleifera is a robust plant crop that grows in the tropics and subtropics. It can thrive under poor or moderately dry soils and grow up to 15 meters tall. The trunk is usually 20-40cm in diameter. Moringa is universally known for its pharmacological action and is utilized not only in folk medicine but also in recent research. The leaves and seeds of the plant are especially renowned for their inexpensive and eco-friendly bio sorption properties. Furthermore, it has been illustrated in research that Mango leaves, upon chemical activation, have greater metal adsorption values compared to some standard adsorbents. This improves their potential not only as coagulants but also as heavy metal and other contaminants removing agents from industrial wastewater.

2.5 Methodology



3. Results and Discussion

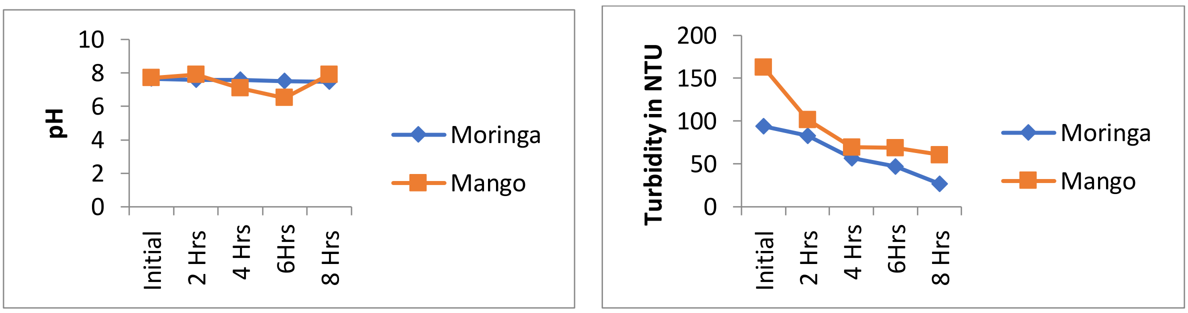


Fig. 5: Variations of pH and Turbidity for the treatment of Raw Pharmaceutical wastewater sample 1 and 2 by using the Natural coagulants Moringa and Mango leaf powdered.

The pH value recorded for the treatment of Moringa leaves shows a generally even trend over the contact duration. In spite of the decrease in pH with a rise in hydraulic retention time (HRT), the decreasing trend is neither sudden nor acute. This implies that the concentration of active ingredients present in the Moringa extract is quite consistent with time and small variations. The pH consistency implies that Moringa leaves have a relatively consistent chemical environment during the treatment process. The pH trend for Mango leaves is different. The pH values show extreme variation during the initial phases, possibly due to differential release of the organic constituents or with the wastewater matrix. However, after reaching the 6-hour HRT mark, there is a significant increase in pH levels. This upward trend in pH, especially evident after the 6-hour period, may be attributed to changes in the chemical composition of the Mango leaf extract or microbial activity that leads to a shift toward alkalinity. Such a trend could indicate the formation of alkaline by products or the reduction of acidic components over time. When examining the turbidity levels for both natural coagulants, a clear trend of reduction is observed throughout the treatment duration. This decrease in turbidity implies that the suspended solids or particulate impurities present in the wastewater are being effectively removed. Specifically, the application of Moringa leaves results in a turbidity reduction of approximately 71.27%, while Mango leaves achieve a slightly lower, but still substantial, reduction of about 62.57%. These results, illustrated in Figure 5, highlight the effectiveness of both natural coagulants in reducing the turbidity of pharmaceutical wastewater over time, supporting their potential role in eco-friendly wastewater treatment processes.

Fig. 6: Total solids and Total Dissolved solids for the treatment of Raw Pharmaceutical wastewater sample 1 and 2 by using the Natural coagulants Moringa and Mango leaf powder

In reduction of Total Solids (TS) concentration from pharmaceutical wastewater treated by natural coagulants, a clearly defined trend is seen with the use of Moringa leaves. Total solids show continuous and consistent reduction with time. Such continuous reduction shows that effective and continuous removal of suspended particulate matter takes place, which is perhaps due to efficient settling or degradation of solid particles during treatment. Such continuous reduction of total solids by Moringa leaves is due to the natural coagulative nature of the leaves, which allows efficient sedimentation by particle agglomeration and bridging mechanisms. When Mango leaves are employed as a natural coagulant, the trend for total solids is not the same. The concentration at the start is comparatively higher than Moringa. The total solids show small fluctuations with time, but with a well-defined peak at the 4 to 6-hour hydraulic retention time (HRT). Such a peak is due to retarded coagulation or intermediate particle interactions. Following such a peak, a small reduction in concentration of total solids can be seen at the 8-hour HRT, showing continued improvement in efficiency of settling. Such a trend for Mango leaves is such that the concentration of total solids is comparatively constant with small fluctuations, showing less efficient removal compared to Moringa.

The relatively superior performance of Moringa leaves to decrease the concentration of the total solids is due to their higher sedimentation ability. This is due to the natural coagulant proteins and other active molecules in them that are more contributory to the coagulation and flocculation process than the Mango leaves. With a keen eye on the Total Dissolved Solids (TDS), even the treatment efficiency shows stark contrasts between the two natural coagulants. For the Moringa leaves, the TDS is relatively more stable throughout the treatment, except for a decrease by the 8-hour HRT. This indicates that while Moringa reduces suspended solids, its efficiency over the dissolved solids is minimal. This indicates that the majority of the dissolved constituent remains in the solution, and the TDS profile is relatively stable throughout the test.

On the contrary, when Mango leaves are utilized as the natural coagulant, the TDS concentration begins at a lower initial concentration of about 632 mg/L. There are progressive changes in the TDS concentrations with a steep increase or peak at the 4 to 6-hour HRT. This is perhaps because half of the soluble organic substances or salts from the Mango leaves are released upon reaction with wastewater. Gradually, there is a moderate decline by the 8-hour HRT, indicating a partial stabilization of the dissolved solid concentration. The changes in the behavior of TDS are diagrammatically depicted in Figure 6, showing the observation that Mango leaves act differently with dissolved solids with Moringa leaves. Overall, these findings depict the better performance of Moringa leaves in removing total solid content, while both the coagulants have no measurable but finite effects on dissolved solids in pharmaceutical wastewater treatment.

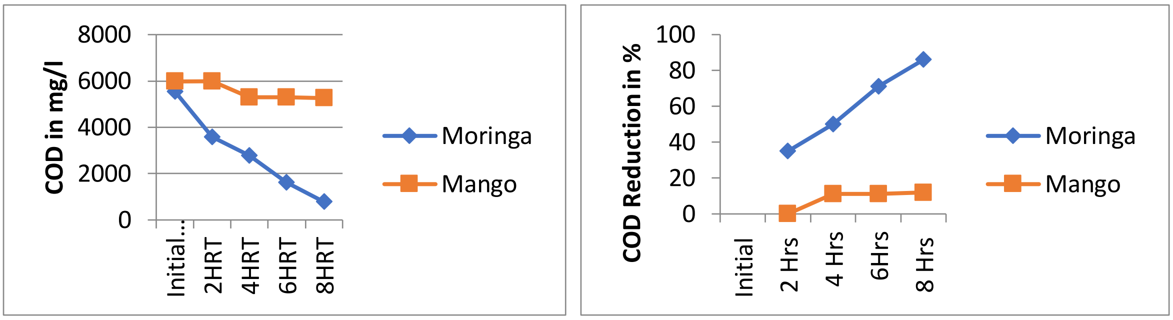


Fig. 7: Variations of COD for the treatment of Raw Pharmaceutical wastewater sample 1 and 2 by using the Natural coagulants Moringa and Mango leaf powder

Variation of Chemical Oxygen Demand (COD) concentration with time is extremely insightful on the performance of natural coagulants in treating the pharmaceutical wastewater. Where Moringa leaves are used as the coagulant, the initial COD concentration of 5547 mg/L undergoes a remarkable and consistent decrease with time during the treatment. At the last 8-hour hydraulic retention time (HRT), COD concentration drops as low as 781 mg/L. Such a sharp drop corresponds to a remarkable removal efficiency of as high as 86%. The consistent decrease of COD concentration throughout the treatment time is a clear indication that Moringa leaves allow active and sustained degradation of organic impurities in the wastewater. The treatment performance of natural coagulants is Shown in Table 2.

Table 2: Treatment Performance of Natural Coagulants (Moringa and Mango Leaves) on Pharmaceutical Wastewater

Parameters	Sample 1(Raw)	Sample 2(Raw)	Sample 1 treated with NC Moringa Leaf Powder	Sample 2 treated with NC Mango Leaf Powder
Turbidity	94 NTU	163 NTU	21 NTU	61 NTU
COD	5547 mg/l	5977 mg/l	781 mg/l	5258 mg/l
Turbidity Reduction %	-	-	77.65	62.6
COD Reduction %	-	-	86	12

However, the trend of performance of the application of Mango leaves as a coagulant is very different, though. The COD concentration of wastewater treated with Mango leaf extract is much higher at 5977 mg/L. At 8-hour HRT, the reduction is very low, with the COD concentration being reduced only marginally to 5258 mg/L. This is only 12% removal efficiency, much worse compared to that of the treatment performance of Moringa leaves. The absence of reduction indicates that Mango leaves do not possess so much coagulation or organic degradation ability when applied in the same conditions. The superior performance of Moringa leaves can be attributed to the cationic polymers naturally present in the leaf extract. The polymers combine with anionic pollutant particles in the wastewater, effectively neutralizing the charges. The charge neutralization brings about a bridging effect whereby particles combine to create larger flocs. As a result, the flocs settle more easily, increasing the removal of suspended and dissolved organic matter from the solution. The enhanced floc formation and subsequent sedimentation facilitate better COD reduction and overall purification of the wastewater (Nath et al. 2021).

Furthermore, when observing the COD reduction trend over different time intervals, Moringa leaves show a consistent and progressive improvement. At 2 hours HRT, the COD reduction is already noticeable at around 40%. This efficiency continues to improve steadily over time, reaching nearly 90% by the 8-hour mark. This trend underscores the time-dependent efficiency of Moringa as a natural coagulant in breaking down organic matter and improving water quality. On the other hand, the reduction percentage observed for Mango leaves remains relatively low throughout the treatment period. Initially, the COD reduction is negligible. A slight increase is observed at the 4-hour HRT, where the reduction percentage rises modestly. However, beyond this point, the COD removal remains nearly stagnant, stabilizing between 15% to 20% up to the 8-hour HRT. This minimal improvement suggests that Mango leaf powder lacks the necessary coagulative and reactive properties required for significant organic matter degradation in wastewater.

In conclusion, the comparative analysis clearly demonstrates that Moringa leaves are far more effective than Mango leaves in reducing COD levels in pharmaceutical wastewater. The higher efficiency, stable performance over time, and superior coagulation mechanisms of Moringa make it a promising and eco-friendly alternative for wastewater treatment applications.

4. Analytical Procedure

pH

pH ,Turbidity, Total Solids (TS),Total Dissolved Solids (TDS) and Chemical oxygen Demand (COD) were measured based on the Standards methods.

Total Solids (TS)

The raw sample was collected and evaporated in a weighing dish (crucible) and dried to a constant weight in an oven at 105°C and the Total Solids of the sample is obtained using

$$TS\ (mg/L) = (w2-w1)/\ \text{Volume of sample}$$

Where, w1 = empty weight of crucible

w2 = (weight of crucible + residue).

Total dissolved solids (TDS)

For determining Total Dissolved solids (TDS), remains of the sample was filtered through a glass fibre filter (1μm, Whatman GF) and subjected to evaporation at 105°C, for one hour. The TDS is then obtained using

$$TDS\ (mg/L) = (w2-w1)\ /\ \text{Volume of sample}$$

Where, w1 = Empty weight of crucible

w2 = (weight of crucible + filtrate residue).

Chemical Oxygen Demand (COD)

COD represents the amount of oxygen required to oxidize all organic compounds both biodegradable and non-biodegradable present in the wastewater to carbon dioxide and water. The determination of COD typically takes 2 hours and involves the digestion of organic matter using strong oxidizing agents in a COD digester apparatus. Potassium dichromate ($K_2Cr_2O_7$) is the preferred oxidizing agent for this process. The sample is cooled to room temperature, and the excess dichromate is titrated with ferrous ammonium sulfate (FAS). The amount of oxidizable organic matter, expressed as an oxygen equivalent, is then calculated as the COD using the following formula:

$$\text{COD (mg/l)} = [(\text{A-B}) \times \text{molarity of FAS} \times 8000 / (\text{Volume of sample})]$$

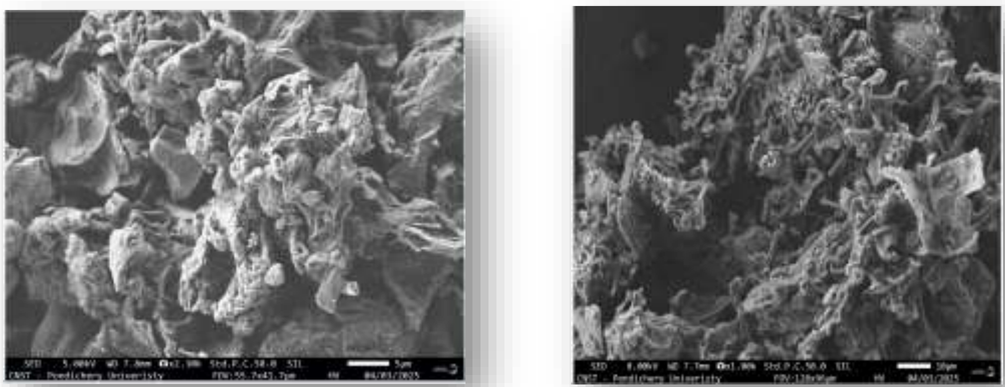
Where, A = titrant value of blank solution; B = titrant value of the sample.

Chemical Oxygen Demand (COD) Reduction

Percentage of COD reduction is commonly calculated by using the formula

$$\%\ \text{COD Reduction} = \frac{COD\ Inlet - COD\ Outlet}{COD\ Inlet}$$

5. FE SEM –EDX analysis of various elements present in Moringa leaves (Moringa oleifera).

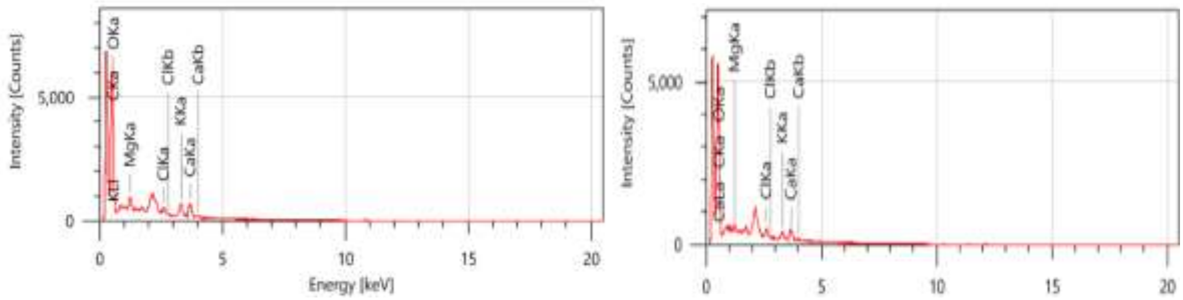


(a) (b)

Fig. 8: FE SEM image of Moringa Leaves (a) before and (b) after Treatment with Industrial Pharmaceutical wastewater

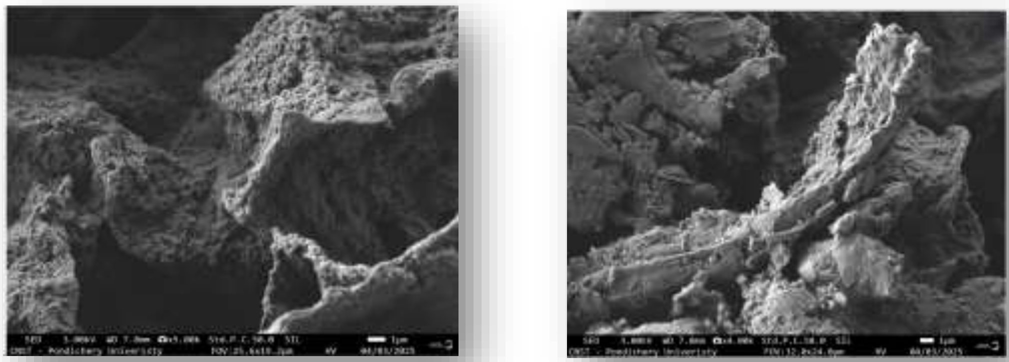
Figure (a) illustrates the microstructural morphology of Moringa oleifera leaf powder prior to treatment**.** The image reveals a distinctly porous and irregularly folded surface**,** characterized by numerous crevices and voids. These structural features contribute to a large specific surface area**,** which plays a crucial role in enhancing the adsorptive and coagulative efficiency of the natural coagulant. The presence of folded, flake-like layers provides multiple reactive sites for pollutant binding, particularly aiding in the removal of suspended solids and organic contaminants from wastewater. Such morphological traits are indicative of an effective natural material for coagulation applications.

Figure (b)**,** offering a broader perspective of the particle aggregation behavior of Moringa leaf powder. The image shows the interconnected and clustered nature of particles, which appear to be intertwined and fused together. This aggregation is likely influenced by the presence of natural biopolymers such as proteins and polysaccharides**,** inherently present in the leaf material. These biopolymers act as bridging agents, promoting particle agglomeration, floc formation, and enhanced sedimentation. The observed macrostructural organization further supports the functional role of Moringa leaves in efficient pollutant removal through sedimentation and flocculation mechanisms. This image as supporting evidence of macro-scale structure formation, contributing to the effective settling of solids hence there is reduction in Turbidity and COD.



(C) (d)

Fig. 9: EDX analysis of Moringa Leaves (c) before and (d) after Treatment with Industrial Pharmaceutical wastewater

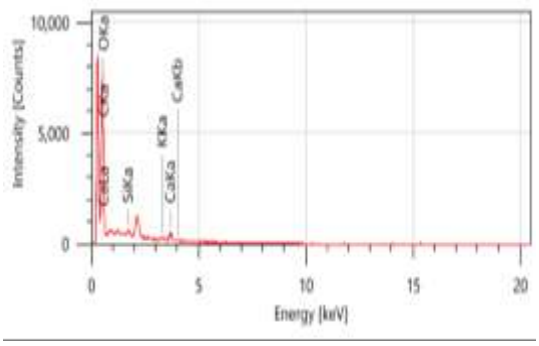


(e) (f)

Fig. 10: FE SEM image of Mango Leaves before (e) and after (f) Treatment with Industrial Pharmaceutical wastewater

Figure (e) illustrate the surface of the Mango leaf powder before treatment appears porous with irregular ridges and cavities**,** featuring loosely packed granules and a relatively rough morphology. These surface characteristics suggest a high degree of surface area**,** which may facilitate adsorption and pollutant interaction during the initial treatment stages. The presence of porous microstructures and surface voids supports the potential of Mango leaves to act as a natural coagulant. The irregular surface offers active binding sites for suspended impurities and organic compounds. However, the structure appears less fibrous and compact compared to Moringa leaves, which may limit its overall coagulative performance. Mango leaf powder exhibits a porous structure, its coagulation efficiency is moderate, potentially due to lower bioactive compound density or weaker charge neutralization mechanisms compared to Moringa.

Figure (f) shows significant structural changes**,** including the presence of layered, compacted residues and clustered particulates. Some regions exhibit a gel-like coating or film, likely formed by adsorbed organic and inorganic contaminants. The central fibrous structure in the image may indicate binding of pollutants onto the leaf matrix. The changes suggest that the Mango leaf surface has interacted with pollutants during the coagulation process, leading to partial agglomeration and pollutant trapping. However, the extent of aggregation and surface deposition appears less dense or uniform than typically seen in high-efficiency coagulants. This supports experimental findings of limited COD and turbidity reduction using Mango leaves. Mango leaves contribute to pollutant removal**,** but with lower sedimentation and floc formation capacity**.** The fibrous elements visible post-treatment may be remnants of organic compounds or biofilms, but overall, the structural integrity remains less altered compared to Moringa hence there is a lower percentage of Turbidity and COD reduction.



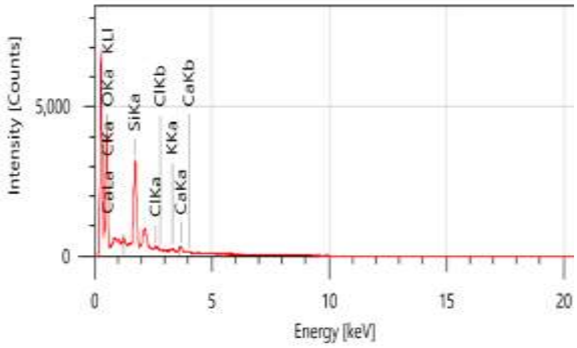


Fig. 11: EDX analysis of Mango Leaves before (g) and after (h) Treatment with Industrial Pharmaceutical wastewater

The Field Emission Scanning Electron Microscopy (FE-SEM) technique was employed to investigate the microstructural characteristics of both Moringa and Mango leaves. This high-resolution, high-vacuum imaging method allows for detailed visualization of surface morphology at magnifications suitable for analyzing natural coagulants (Dey et al. 2022). The morphological changes in Moringa and Mango leaves, both before and after treatment with raw pharmaceutical wastewater samples (Sample 1 and Sample 2), are depicted in Figures 8, 9, 10, and 11 In addition to morphological analysis, elemental composition was examined using Energy Dispersive X-ray Spectroscopy (EDX). For fresh Moringa leaves, the elemental content was found to be: Carbon (C) – 30.84%, Oxygen (O) – 54.89%, Magnesium (Mg) – 1.20%, Chlorine (Cl) – 2.21%, Potassium (K) – 2.57%, and Calcium (Ca) – 4.64% (atom/mol). After treatment with pharmaceutical wastewater, these values shifted to C – 31.78%, O – 57.89%, Mg – 1.80%, Cl – 1.88%, K – 4.73%, and Ca – 5.86%, indicating an increase in elemental uptake. This change reflects the adsorptive potential and active binding capability of Moringa leaves, which effectively removed various ions and organic compounds from the wastewater.

Similarly, Mango leaves were analyzed using EDX. The untreated Mango leaf powder showed elemental values of C – 33.33%, O – 60.73%, Silicon (Si) – 0.78%, K – 1.21%, and Ca – 3.95%. Post-treatment, the elemental composition changed to C – 41.86%, O – 42.49%, Magnesium – 0.97%, Si – 10.82%, Cl – 0.71%, K – 0.87%, and Ca – 2.29%. These shifts demonstrate that Mango leaves also exhibit notable adsorption and elemental exchange, suggesting their suitability as a natural coagulant for pharmaceutical wastewater treatment.

However, despite their absorptive properties, Mango leaves demonstrated lower efficiency in reducing Chemical Oxygen Demand (COD) when compared to Moringa. This difference is attributed to the less effective flocculation, charge neutralization, and organic matter binding capacity of Mango leaves. Thus, while both materials show promising potential as eco-friendly coagulants, Moringa leaves offer superior performance in terms of COD reduction and overall treatment efficiency.

6. Conclusion

The samples were collected from a pharmaceutical industry, exhibiting different characteristics, with turbidity levels of 94 NTU and 163 NTU for Sample 1 and Sample 2, respectively. The initial COD values were 5547 mg/L for Sample 1 and 5977 mg/L for Sample 2,. When treated with plant-based natural coagulants, like Moringa leaf powder (Moringa oleifera) and Mango leaf powder (Mangifera indica) the wastewater showed a turbidity reduction of 77.65% and 62.6%, respectively. The COD reduction achieved was 86% with Moringa leaves and 12% with Mango leaves. Carbon-based compounds present in pharmaceutical wastewater can significantly increase the COD concentration. The organic elements, such as carbon (C) and oxygen (O), adsorbed by Moringa leaves as confirmed by FE-SEM analysis contribute to the reduction of COD concentration. Although mango and peepal leaves adsorb elements like C, Si, K, Mg, Cl, Ca, and Al, Al and Mg are considered organic metals. The remaining elements (C, Si, K, and Ca) are inorganic and do not contribute to COD reduction. Therefore, Moringa leaves, as a natural coagulant, show the highest percentage of COD concentration reduction. Although the treated wastewater does not fully meet the permissible limits set by the Pollution Control Board, the treatment significantly reduces COD and other parameters from their initial levels. Compared to conventional chemical methods, this plant-based approach offers a relatively high reduction in turbidity and COD at a lower cost, using easily available materials with better efficiency. FE-SEM and EDX analyses were conducted to examine the microstructural morphology of Moringa and Mango leaves before and after treatment with industrial pharmaceutical wastewater. These analyses also identified the presence of essential elements such as C, O, Mg, Si, Cl, K, and Ca in the natural coagulants. Since these elements are beneficial for cultivation, the bottom-settled sludge can be potentially utilized as a fertilizer.

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