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Research Article

A Synergistic Effect of *Moringa oleifera*-Based Coagulant and Ultrafiltration for the Wastewater Treatment Collected from Final ETP

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Provision of safe drinking water, devoid of aetiologies, is an all-time challenge due to the usage of unsafe chemicals in most of the water treatment processes. The main objective of the present paper is to evaluate the use of *Moringa oleifera* (MO) as a natural coagulant in coagulation/flocculation (C/F) followed by the ultrafiltration (UF) of Final Effluent Treatment Plant wastewater treatment which can also be employed as an alternative to the present conventional methods of treatment. Process efficiency was evaluated in terms of chemical oxygen demand (COD), biochemical oxygen demand (BOD), turbidity, total hardness, alkalinity, ammoniacal nitrogen, and zeta potential along with permeability and fouling behaviour of the membrane. A significant improvement in both the physical and chemical characteristics of the effluent quality is showing a clearer colour and a greater reduction in BOD (89.74%) and COD (63.80%) values, while pH was in the acceptable range for effluent disposal. The results indicate a lower membrane fouling rate (49%), an increase in permeate flow, and better quality of the permeate, proving that the C/F-UF treatment is an effective and efficient technique for wastewater treatment. Eventually, the treated wastewater obtained with this process generates better quality water and preserves the aquatic ecosystem.

1. Introduction

Water quality and accessibility have always been critical factors in defining how and where individuals may inhabit and also their standard of living. Despite the fact that there is indeed a plethora of fresh water on the planet, it may not always be accessible whenever and how it is desired, nor has it always been of desirable quality for all needs. Lack of safe drinking water is a major global issue and a leading cause of morbidity and mortality. According to the World Health Organization (WHO), 785 million individuals globally do not have access to potable water, and water-borne

pathogens cause 485,000 deaths annually [1]. The sustainability of existing freshwater resources is being impacted by elements such as rapid urban growth, expanded agricultural operations, chemical use, soil depletion, dense population, and inadequate waste management [2]. Toxic and nondegradable waste generated by medicine, textiles, dyeing, and other enterprises makes a significant contribution to not only a greater incidence of water-borne diseases caused by resistant bacterial strains in drinking and surface waters but also to physiological health implications such as genetic disorders, cancer, and neurological problems. Such toxins also reduce the turbidity and oxygen concentration of water, lowering ecosystem productivity and exposing marine communities to risk [3–8].

Conventional wastewater treatment processes entail major difficulties in removing contaminants. The use of chemical agents in wastewater treatment, such as aluminium sulphate, chlorine, potassium permanganate, ferric sulphate, polyethylene terephthalate (PET), which inadvertently lead to various major health concerns when used over extended periods of time, is indeed a subject of discussion [9–12]. Furthermore, because of their massive carbon load and high energy consumption, they are not reliable. Depending on the treatment method, these activities produce a large quantity of greenhouse gas emissions each year, ranging from 61 to 161 kg of carbon dioxide equivalent for every population equivalent (CO_2eq/PE). Their yearly energy usage might range between 15 and 86 kWh/PE [13]. As a result, existing treatment procedures are both costly and inadequate to meet the required compliance standards [14]. As a response, innovative methods must be created which incorporate environmental and socioeconomic aspects concerning sustainable development while reducing by-products, improving physicochemical characteristics, and using fewer resources [15].

Coagulation is a conventional treatment technique that is commonly used at the initial stages of effluent treatment processes. Inorganic, synthesized organic, and organic polymers are the various varieties of coagulants [16]. Chemical coagulants like aluminium sulphate and ferrous sulphate are generally employed in the treatment processes. The use of chemical coagulants in water treatment plants has been linked to both people and ecological health concerns. The generation of a large volume of sludge by chemical coagulants calls for the need to make use of natural-based coagulants which have a variety of benefits, including reducing costs, preventing variations in the pH of the treated water, reducing the production of sludge, and providing greater biodegradability [17, 18]. In this scenario, the seed extracts/powders of a tropical multipurpose tree, *Moringa oleifera* (MO), are being used in several countries either to clean drinking water or to treat wastewater [19–22]. The proteins or chemical components present in MO possess coagulation, antimicrobial, and pollutant removal activities, thereby making it a convenient natural agent for water treatment [23–25]. Crude extracts from different tissues of MO have been analyzed that show antibacterial activity against both gram-negative and gram-positive bacteria [26]. However, due to the characteristics of Final Effluent Treatment Plant (FETP) wastewater, the physicochemical step of coagulation/flocculation alone is not enough to remove all the pollutants necessary to meet the standards for reuse or releasing the treated wastewater into receiving water bodies, so the study of a subsequent step is required [27]. Membrane processes, which include reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF), and microfiltration (MF), have been widely adopted for tertiary treatment and for the purification and reuse of secondary effluents [28]. Ultrafiltration shows promising application potential with several advantages, such as high product quality and easy handling [29, 30]. The goal of this work is to devise an integrated treatment process for FETP wastewater comprising of C/F-UF technique.

2. Materials and Methods

2.1. Wastewater Sampling and Characterization. Final Effluent treatment Plant is located (21°37′03″ N 72°58′54 ″ E) in Ankleshwar, Gujarat. The treatment plant receives complex treated waste from Ankleshwar, Jhagadia, and Panoli Industrial Estates and coveys the treated water through a 52.76 km pipeline into the Arabian Sea. It is well established that the FETP, from its inception to date, has never performed as per the prescribed norm set by the Gujarat Pollution Control Board [31]. In the present study, the wastewater sample was collected from the outlet of the treatment plant in a clean polyethylene bottle following a standard regime. The sample was homogenized, fractioned, and suitably stored under refrigeration for subsequent use. Before the analytical procedures, the sample was characterized for physicochemical parameters: pH, turbidity, chemical oxygen demand (COD), biochemical oxygen demand (BOD), ammoniacal nitrogen, alkalinity, total hardness, pH, sulphate, and also zeta potential. Zeta potential was measured in the coagulant and wastewater during the coagulation/flocculation process. The zeta potential is closely related to the surface charges of a colloidal system and provides an indication of the stability of colloidal particles [29]. All experiments to determine the physicochemical parameters were performed in triplicate and followed the methodology of the Standard Methods for the Examination of Water and Wastewater [32]. The initial physicochemical characteristics of selected wastewater are presented in Table 1.

2.1.1. Estimation of Turbidity and Zeta Potential. pH was noted in situ using pHTestr 20. Turbidity was evaluated using a systronic double beam spectrophotometer 2203 at 425 nm [33]. Zeta potential was recorded using dynamic light scattering (Model: NPA152-31A-0000-000-90M, Make: Metrohm).

2.1.2. Estimation of COD by Open Reflux Method. In a digestion tube, 10 ml sample, 5 ml potassium dichromate, and 15 ml silver sulphate (1 gram of silver sulphate/100 ml of sulphuric acid) were slowly combined together. A pinch of sulphamic acid and 0.2 grams of mercuric sulphate were also added. The sample was thoroughly homogenized. On a COD digestion unit, the material was exposed to open reflux digestion for 2 hours at 150°C. Following digestion, the sample was cooled to room temperature and an equivalent

Table 1: The physicochemical analysis of wastewater and discharge standards (CPCB and BIS, 2012).

Parameter	Unit	WW	Inland surface water (CPCB)	Marine coastal areas (CPCB)	Drinking water (BIS, 2012)
pH	—	6.9	5.5 to 9.0	5.5 to 9.0	6.5-8.5
Turbidity	NTU	35.0	—	—	1
Chemical oxygen demand (COD)	mg/l	585.66	250	250	—
Biochemical oxygen demand (BOD)	mg/l	282.66	30	100	—
Ammoniacal nitrogen	mg/l	21	50	50	0.5
Alkalinity	mg/l	2000	—	—	200
Total hardness	mg/l	1824.66	—	—	200
Zeta potential	—	-6.47	—	—	—

amount of distilled water was added (30 ml). The sample was titrated with 0.1 N ferrous ammonium sulphate (FAS) and 1-2 drops of ferroin indicator. The colour changed from bluish green to reddish brown. S is the volume of titrant used to titrate the sample. The same approach was also used for blank (B) [32].

$$\text{COD mg/l} = \frac{(B - S) \times N \times 8 \times 1000}{\text{sample (ml)}}, \quad (1)$$

where N = normality of FAS; 8 = milliequivalent weight of oxygen.

2.1.3. Estimation of BOD by Alkali-Azide Method. Two BOD bottles (300 ml) were filled with the sample in situ. One BOD bottle was fixed at 20°C for 5 days (DO_f), while the other BOD bottle was used to determine the initial DO (DO_i). To prevent bubbling, manganese sulphate (1 ml) and alkali-iodide-azide (1 ml) were injected underneath the lower meniscus of the sample. By rotating the bottle, appropriate mixing was attained, and brown precipitate build-up was observed. The precipitate was allowed to settle before being dissolved in 1 ml of H_2SO_4. The sample (100 ml) was titrated with 0.025 N sodium thiosulphate and freshly prepared starch (4-5 drops) as an indicator. The sample changed from blue to colourless, and the final burette reading was noted as S. Same procedure was followed for DO_f to calculate BOD [32].

$$\text{Dissolved Oxygen mg/l} = \frac{S \times N \times 8 \times 1000}{V_2(V_1 - V)/V_1},$$
$$\text{BOD (mg/l)} = (DO_i) - (DO_f), \quad (2)$$

where N = normality of sodium thiosulphate; V = ml of $MnSO_4$ and alkali-iodide-azide (2 ml); V_1 = BOD bottle volume (300 ml); and V_2 = titrated volume of the sample (100 ml).

2.1.4. Estimation of Ammoniacal Nitrogen by the Spectrophotometric Method. Sample (100 ml) was adjusted for pH (10.5) by adding the required amount of zinc sulphate and sodium hydroxide. The precipitate was filtered using Whatman no 42. The mixture was treated with a drop of EDTA and 3 ml of Nessler's reagent. The sample was thoroughly mixed, and the absorbance was recorded at 410 nm after 10 minutes. Simultaneously, a reading for blank (distilled water) was also recorded [34].

2.1.5. Estimation of Alkalinity by Sulphuric Acid Titration Method. Sample (25 ml) was mixed with 2–3 drops of phenolphthalein indicator. If a pink colour appears, titrate the mixture with the titrant (0.02 N sulphuric acid (H_2SO_4) and record the burette reading. Furthermore, phenolphthalein alkalinity is missing if the pink colour does not appear (in the present study, phenolphthalein alkalinity was absent). Following the titration, 2-3 drops of methyl orange were added to the sample, and the colour of the mixture changed from yellow to orange. S represents the volume of titrant consumed [32].

$$\text{Alkalinity } (CaCO_3 \text{mg/l}) = \frac{S \times N \times 50 \times 1000}{\text{sample (ml)}}. \quad (3)$$

2.1.6. Estimation of Total Hardness by EDTA Titration Method. To 50 ml of sample, 2 ml of ammonia buffer and an inhibitor were added. The sample was titrated with 0.01 M ethylenediaminetetraacetic acid (EDTA) and Eriochrome Black-T (3-4 drops) as an indicator. The colour changed from wine red to blue and was noted as S. The same procedure was followed for the blank (distilled water) and recorded as B [32].

$$\text{Total Hardness } CaCO_3 \text{mg/l} = \frac{(S - B) \times C \times 1000}{\text{sample (ml)}}, \quad (4)$$

2.2. Collection and Characterization of MO Seeds. The MO seeds were collected from a local market situated in Gandhinagar, Gujarat, and stored under ambient laboratory conditions with temperatures varying from 20 to 28 degree Celsius. Further, the morphological and qualitative characterization of the seeds was studied using a standard protocol (Table 2). The seed's length and diameter were examined with a 1 mm precision tape and a digital vernier (0.01 mm precision) [35]. The weight of the seed was recorded in grams using a digital weighing scale. The coat and wings of Moringa oleifera seeds were manually removed. The seeds were dried, crushed to a fine powder, and sieved through a 44 mm sieve. Soxhlet system was used to extract the oil. 10 grams of powdered MO seeds was treated with 210 ml of

Table 2: The morphological and qualitative characterization of MO seeds.

Characteristics	Indian MO seeds
Shape	Globular
Colour	Dark brown
Diameter (cm)	1
Length (cm)	1.5-2
Average weight (g)	0.3
Oil (%)	32.4%
Protein (%)	27.8%

ethanol for 6 hours. The solvent was evaporated in a rota vapour [36]. Protein estimation was conducted as per Liang et al. Degreasing of powdered seeds was done using ethyl acetate for 48 hours (ratio of liquid to material 1 : 6 mg/ml). Tris-HCl extraction buffer was added to the sample and incubated for 100 mins at room temperature. After incubation, the sample was centrifuged at 3,000*g* for 15 minutes. The supernatant was subjected to purification using ammonium sulphate precipitation method and desalting. The final product was freeze-dried at −60°C for 24 hours [37].

2.3. MO Coagulant and Ultrafiltration Apparatus. Extraction of seed oil was performed using an ethanol extraction process wherein powdered seeds were mixed with ethanol for 30 minutes on a magnetic stirrer. The sample was centrifuged at 5000 rpm for 10 minutes to segregate the residues, whereupon the supernatant was decanted. The residue was dried for 24 hours at room temperature [38]. Coagulant preparation was as per Ndabigengesere and Narasiah procedure [39]. The dried residue was weighed and mixed with the correct amount of distilled water. Following multiple experiments ranging from 0.5 to 10%, a concentration of 5% was employed throughout the study. The solution was mixed for 10 minutes at 150 rpm with a magnetic stirring and allowed to stand for twenty minutes. Before being transmitted through a 0.45 mm membrane, the suspension was filtered using a Whatman number 42. To avoid qualitative deterioration and efficacy decline due to preservation, the supernatant was used as a coagulant the very same day. The membrane filtration was carried out using a polyether sulfone (PES) hollow fibre ultrafilter with a molecular weight cut-off (MWCO) of 100 kDa. Thickness of the membrane was 0.12 mm, and the contact angle was 60 degrees. A feed tank, peristaltic pump for filtering, pressure gauge, and collector are all included in the system as shown in Figure 1.

2.4. Experimental Protocols

2.4.1. Pretreatment Coagulation/Flocculation Experiment: The Study Was Conducted in a Designed and Developed Reactor. The reactor is made up of an acrylic sheet of rectangular shape having dimensions of 15 cm × 15 cm × 25 cm with an automatic stirrer attached to it as shown in Figure 2. The reactor was filled with 2 litres of wastewater and agitated for rapid mixing. During the process, an appropriate dosage of coagulant was added using Eppendorf pipettes. The following requirements have been used for coagulation/flocculation: a fast-mixing speed of 100 rpm for 2 minutes, a steady mixing speed of 50 rpm for 10 minutes, and a settling period of 60 minutes [40]. After that, the supernatant was separated for filtration studies and quality assessment.

2.4.2. Ultrafiltration Experiment. The durability of the membrane module was checked before each UF investigation by monitoring clean water permeability at 20 degree Celsius. Thereafter, the feed reservoir was filled with the sample after MO coagulation. The process was operated at a constant pressure of 5 bar, temperature of 25 ± 1°C, and constant velocity of 1 m/s. In order to determine the efficiency of the ultrafiltration process, the filtrate was assessed for its physicochemical characteristics and 84% sample recovery was recorded at the end of the experiment.

2.4.3. Membrane Permeability and Fouling Characterization. Ultrafiltration of the sample took approximately 60 minutes, plus 60 seconds of forwarding flushing with deionized water. The quantity of infiltrate was determined in order to calculate the permeation using [41]

$$Pm = \frac{Vp}{(\Delta p \times a \times \mathrm{tp})}, \tag{5}$$

where Pm represents the permeability ($\mathrm{L \cdot m^{-2} \times h^{-1} \times ba\ r^{-1}}$), Vp is the permeated volume in litres (l), A stands for membrane surface area ($\mathrm{m^2}$), tp stands for permeate collection time collection (hr), and p refers to transmembrane pressure (bar).

Deionized water fluxes (Jw) were calculated using formula (6) in this investigation to determine membrane fouling (M_f) prior to (Jw_i) and after (Jw_f) ultrafiltration.

$$\%M_f = \frac{\left(\mathrm{Jw}_i - \mathrm{Jw}_f\right)}{\mathrm{Jw}_i \times 100}, \tag{6}$$

3. Results and Discussion

3.1. Oil Extraction from MO Seeds. Oil extraction of MO seed is encouraged to improve the wastewater treatment process efficacy. The electrostatic patch phenomenon, which is indeed a surface mechanism, is used by MO to reduce turbidity from wastewater [42]. The seed's oil content might create an emulsion or film coat, which could prevent interaction with the reactive surfaces and thus limit floc production. As a response, oil extraction might improve turbidity removal, resulting in improved coagulation-flocculation [43]. MO seeds were processed for oil via ethanol extraction. In general, the oil content of MO seeds is around 35-40% [44]. In the present investigation, approximately 16.32 percent of oil was recovered from MO seeds.

3.2. Coagulation/Flocculation by MO Seed Powder. The present research examined the usage of MO seed coagulant in FETP wastewater treatment. MO seeds proved effective in improving the physicochemical water quality characteristics

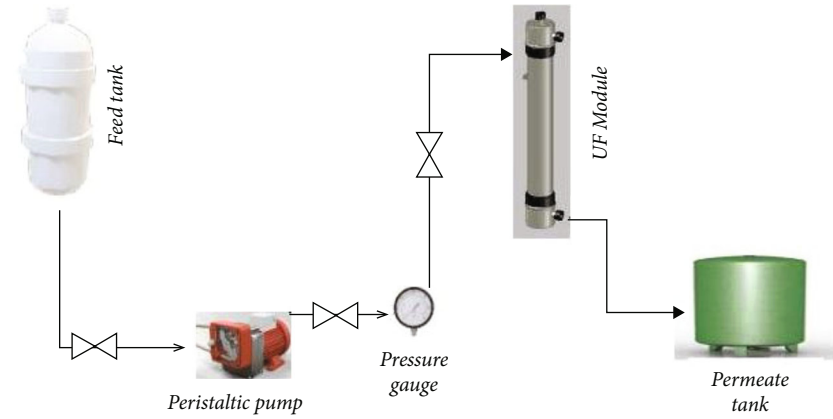

Figure 1: Schematic diagram of ultrafiltration set-up.

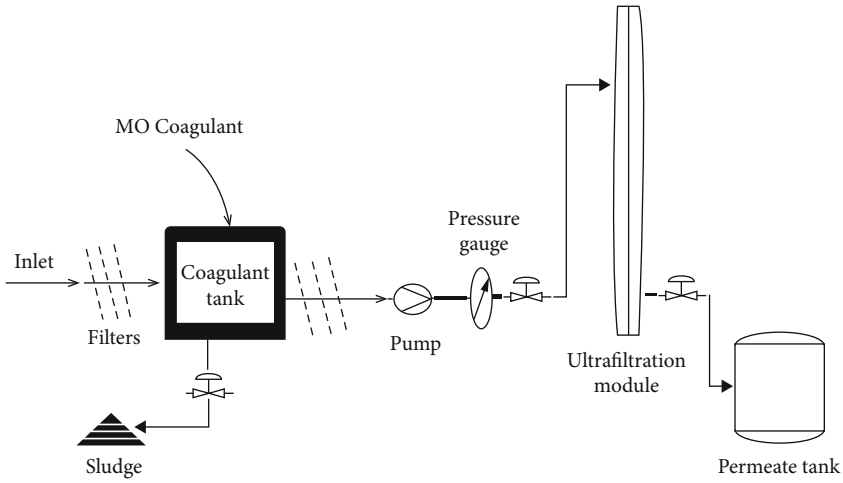

Figure 2: Flow chart of natural coagulation and ultrafiltration-based wastewater treatment technology.

of wastewater, as evidenced by the results. The outcomes of the application of MO seeds are summarised in Table 3. The pH of the sample ranged from 6.5 to 8.5. This demonstrated that MO seed had no influence on the pH of the sample, no subsequent steps are necessary to correct the pH values, and also that the coagulant is effective in treating FETP wastewater [45, 46]. 64% reduction in turbidity was observed after the coagulation/flocculation step using MO seed. According to Nkurunziza et al., MO can be used as a coagulant in water treatment on a domestic and industrial scale [47]. Since MO is much more efficient at high levels of turbidity, its widespread use could be especially advantageous even during monsoon season, when water turbidity is at its peak and treatment plants are temporarily closed [48]. High levels of COD (585.66mg/l) and BOD (282.66mg/l) in the wastewater marks the presence of higher load of inorganic and organic matter. MO seeds were found effective in reducing the COD and BOD levels of the sample by 38% and 58%, respectively [49, 50]. However, a dramatic reduction in BOD was observed during the process [51]. Adsorption and charge neutralization is the most likely mechanisms by which the pollutants in wastewater samples were removed by the MO [52]. Ammonia was not removed effectively by the MO seeds during the process, this might be attributed to the fact that MO is a cationic coagulant and could not attract the positive charge of ammonium [53]. A decrease in alkalinity from 2000 mg/l to 1020 mg/l and total hardness from 1824.66 mg/l to 927 mg/l can be observed. Water-soluble, positively charged proteins present in MO seeds could be responsible for the adsorption of alkalinity and total hardness from the sample [54, 55]. The values for zeta potential showed a significant increase during the coagulation-flocculation process, which remained the same for nearly the entire study. After the coagulation/flocculation, electrostatic repulsion reactions occurred during the sedimentation set and altered the suspension stability [56], resulting in the increase of zeta values and demonstrating the load neutralization, since the values remained close to zero [57]. These results indicate that the mechanism involved in the step is charge neutralization [29].

3.3. Ultrafiltration Study. The ultrafiltration process can reduce turbidity to very low levels, but problems such as low removal of dissolved organic matter and low permeate flow can occur. The effluent treated using MO seed was subjected to the ultrafiltration study. As expected, significant

Table 3: Physicochemical characterization of FETP wastewater after the C/F using MO seeds and ultrafiltration.

Parameter	Unit	WW after C/F	% removal after C/F	WW after UF	Total removal (%)
pH	—	6.8	—	6.9	—
Turbidity	NTU	10.08	64	1.1	96.85
Chemical oxygen demand (COD)	mg/l	363.11	38	212	63.80
Biochemical oxygen demand (BOD)	mg/l	118.72	58	29	89.74
Ammoniacal nitrogen	mg/l	19	9.52	15	28.57
Alkalinity	mg/l	1020	49	189	91.50
Total hardness	mg/l	927	49.19	170	90.68
Zeta potential	—	0.19	—	0.21	—

reductions in individual parameters were recorded. A reduction in turbidity from 10.08 NTU to 1.1 NTU was observed which marked overall removal achieved up to 96.85%. Total percent removal of COD and BOD reached 63.80% and 89.74%, respectively, and the final data depicts that the treated effluent complies with the prescribed standard of discharge for the said parameters. Ammoniacal nitrogen was removed only to a small degree in the integrated process (28.57%). Ultrafiltration may offer enhanced reduction in ammonia if wastewater is subjected to pretreatment. In this study, the coagulant used in pretreatment carries positive charge that may have repelled the positive charge on ammonium ion present in the sample. Total removal (%) obtained for alkalinity and total hardness reached 91.50% and 90.685, respectively. The final results correspond to the compliance with the prescribed standard and denote the better efficiency of ultrafilter in the removal of alkalinity and total hardness.

3.4. Membrane Permeability, Fouling Characteristic, and Efficiency of C/F-UF for Wastewater Treatment. The membrane permeability for deionized water was 36.2 L/m2·h·bar. In the case of C/F wastewater, the membrane permeability decreased to 31 L/m2·h·bar. The rate of fouling for the C/F-UF treatment was 49%. Such results can be attributed to the presence of organic pollutants in the sample and the formation of smaller flakes by the C/F process, which can lead to a decrease in the pore diameters and thus reduce permeate passage which causes high rates of fouling.

The C/F-UF process yielded tremendous results for the FETP wastewater treatment in terms of its increased removal efficiency for the parameters, namely, BOD, COD, turbidity, alkalinity, and total hardness. Several authors concluded in their experiments with surface water that the hybrid treatment provides satisfactory results for the removal of organic matter when compared to the filtration process alone, and that the intensity of membrane fouling is intricately related to the type of coagulant used [29, 58]. Direct filtration of samples with high organic matter content, resulting in heavy scale (fouling) on the membrane, makes the organic matter removal difficult, causing irreversible damage to the membrane. Treatment using the combination of ultrafiltration and a coagulation process causes cake formation and polarization of particles present at the membrane surface. This type of fouling on the membrane is reversible and easy to remove by physical methods during filtration. This is consistent with the results obtained by Guo et al. and various others [59–66]. In this sense, it can be said that combined processes result in a lower fouling percentage and significantly greater flow when compared to isolated membrane filtration processes. Therefore, the tested hybrid process can be successfully applied to FETP wastewater treatment, resulting in better water quality and preserving the aquatic ecosystem.

4. Conclusion

The hybrid process systems (C/F-UF) can be effectively employed in the FETP wastewater treatment as compared to the conventional methods. The residue formed in the coagulation-flocculation process is organic. As a result, MO is a great natural coagulant for effluent. MO coagulation did not significantly alter the pH of the water. This appears to be an added benefit over chemical coagulation because it prevents the necessity of pH adjustment after treatment. Permeate quality was improved, and membrane fouling was also reduced significantly. This can be justified by the membrane process' ability to eliminate particles and sediments along with the coagulant's efficacy MOs in the CFS technique. The permeate obtained through this process achieved all of the standards for reuse in different activities. In nutshell, the technology has shown to be cost-effective, environmentally safe, and sustainable, allowing it to be employed in industry as well as many other activities that require better quality water.

Data Availability

All relevant data are included within the article.

Conflicts of Interest

There are no conflicts to declare.

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