

## RESEARCH ARTICLE

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# A Hybrid Coagulation-Microfiltration Approach for Industrial Wastewater Pretreatment

**Anurag Chavan<sup>1†</sup>**, **Sanjay Harke<sup>2</sup>**, **Ravindra Gaikwad<sup>3</sup>** and **Bhavanipant Pande<sup>1</sup>**<sup>1</sup>Department of Environmental Science, School of Basic and Applied Sciences, MGM University, Chh Sambhajinagar, Maharashtra, India<sup>2</sup>Department of Biotechnology, Institute of Biosciences and Technology, MGM University, Chh Sambhajinagar, Maharashtra, India<sup>3</sup>Department of Chemical Engineering, Jawaharlal Nehru Engineering College, MGM University, Chh Sambhajinagar, Maharashtra, India†Corresponding author: Anurag Chavan; [anuragschavan@gmail.com](mailto:anuragschavan@gmail.com)

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## ABSTRACT

Common Effluent Treatment Plants (CETPs) often receive mixed industrial effluents containing high levels of organic and inorganic pollutants, which can overwhelm traditional biological treatment systems. This study investigates the use of a ceramic microfiltration membrane as a pretreatment step to enhance influent quality prior to CETP processing. Composite wastewater samples from various industrial sources were treated through direct membrane filtration and coagulant-assisted filtration with aluminum sulfate and *Moringa oleifera* seed powder. A commercially available membrane (effective area: 0.0407 m<sup>2</sup>) was operated at a transmembrane pressure of 1.0 kg cm<sup>-2</sup>. The membrane's structural and compositional features were characterized using FESEM-EDS, XRD, XPS, and porosity analyses. The results demonstrated significant improvements in the physicochemical quality of the treated wastewater, including reductions in organic content, particulate matter, and stabilization of pH levels favorable for biological treatment. The findings indicate that ceramic microfiltration, combined with chemical and natural coagulants, provides a sustainable and effective method to improve CETP performance and manage industrial wastewater.

## 1. INTRODUCTION

Industrial clusters in emerging economies often produce heterogeneous effluents that are discharged into shared infrastructures known as Common Effluent Treatment Plants (CETPs). These collective facilities offer economies of scale for small and medium-sized industries but frequently face performance challenges when influent wastewater contains organic and inorganic loads that exceed the capacity of conventional biological treatment systems (Padalkar & Hiralal 2018, Singh et al. 2023). Overloading can lead to reduced removal efficiencies, biomass inhibition, excessive sludge production, and increased operational costs. The problem becomes even more complex when the influent consists of mixed industrial wastewater, which varies significantly in chemical composition and pollutant concentrations.

Mixed industrial effluents differ significantly from domestic or single-industry wastewater. They often contain fluctuating levels of chemical oxygen demand (COD), biological oxygen demand (BOD), total suspended solids (TSS), total dissolved solids (TDS), heavy metals, salts, and non-biodegradable compounds.

This variability places a considerable strain on biological reactors, resulting in unstable operation and shock loads that can disrupt microbial activity (Sravan et al. 2024). A field study at an Indian CETP observed a notable decline in reliability and pollutant removal efficiency whenever influent quality fell outside the plant's designed range (Ali et al. 2021). These findings highlight the critical need for robust pretreatment methods to stabilize influent characteristics prior to biological treatment.

Pretreatment stages aim to safeguard downstream biological units by eliminating refractory organics, solids, and colloidal matter, thereby enhancing overall effluent compatibility. Membrane-based separation techniques, particularly microfiltration (MF) and ultrafiltration (UF), have gained recognition as effective methods for removing suspended and colloidal pollutants (Othman et al. 2021). Notably, ceramic microfiltration membranes provide significant advantages over polymeric ones due to their chemical inertness, thermal stability, mechanical robustness, and extended operational lifespan (Hakami et al. 2020, Zhang et al. 2023). These membranes can withstand the harsh conditions often present in industrial wastewater streams, making them well-suited for CETP pretreatment. Recent developments highlight their increasing use in wastewater polishing and hybrid systems, although high capital costs and fouling potential continue to pose challenges (Akash et al. 2024, Dashtban Kenari et al. 2025).

Coagulation–flocculation processes continue to play a vital role in traditional wastewater treatment and have recently been adapted into greener, bio-based methods. Natural coagulants from plant materials have demonstrated promising potential as sustainable alternatives to alum or ferric salts (Ribeiro et al. 2019). *Moringa oleifera* seed powder, in particular, has been extensively researched for its coagulation capabilities; its cationic proteins serve as natural polyelectrolytes that neutralize negatively charged colloids, resulting in the formation of settleable flocs (Ndabigengesere & Narasiah 1998, Yamaguchi et al. 2021). Research indicates that coagulants derived from *Moringa* seeds can significantly reduce turbidity and chemical oxygen demand (COD), providing cost-effective and biodegradable solutions for wastewater treatment (Oladoja & Pan 2015). Recent assessments of their effectiveness in actual wastewater samples, including dormitory and domestic effluents, further validate their practical application (Ogunshina et al. 2023). Combining these bio-coagulants with membrane technologies not only improves pollutant removal efficiency but also decreases membrane fouling, energy use, and sludge toxicity (Alazaiza et al. 2025).

Hybrid coagulation–membrane systems leverage the strengths of both methods: coagulation decreases particle load and membrane fouling, while the membrane offers

effective physical separation of fine particulates and organic substances (Nouira et al. 2025). This complementary interaction enhances filtration efficiency and produces a stable effluent suitable for biological treatment. For CETPs that manage diverse industrial effluents, such hybrid pretreatment approaches are practical and sustainable options for improving influent quality without significant infrastructural changes.

This study examines the feasibility of using commercially available ceramic microfiltration membranes as a pretreatment step before biological processing in a CETP. A composite wastewater from various industrial sources was treated using two methods: direct membrane filtration and coagulant-assisted filtration with aluminum sulfate and *Moringa oleifera* seed powder. The ceramic membrane was characterized through FESEM–EDS, XRD, XPS, and porosity analysis to link material properties with filtration performance. The research aims to assess how effectively ceramic microfiltration, combined with natural and conventional coagulants, can reduce organic, particulate, and dissolved contaminants, thereby enhancing the wastewater's suitability for downstream biological treatment and supporting sustainable CETP operations.

Although hybrid coagulation–membrane systems have been previously studied, their use in treating highly variable composite influent from operating Common Effluent Treatment Plants remains underexplored. This study advances the field in four key ways. First, it assesses treatment performance using real mixed industrial CETP influent rather than synthetic or single-source wastewater, thereby addressing practical variability and shock-loading conditions. Second, it adopts a simultaneous hybrid dosing approach combining aluminum sulfate and *Moringa oleifera* extract to deliberately modify floc characteristics for fouling mitigation, rather than solely targeting turbidity removal. Third, it evaluates fouling reversibility through clean-water flux recovery analysis, offering operational insights into membrane longevity. Finally, it considers the system's role in enhancing downstream biological process stability, positioning the membrane unit as a CETP pretreatment stabilizer rather than merely a polishing step. These aspects collectively illustrate the main contributions of this work.

## 2. MATERIALS AND METHODS

### 2.1. Collection and Characterization of Composite Wastewater

Composite industrial wastewater was collected from the influent channel of a Common Effluent Treatment Plant (CETP) that serves a diverse industrial cluster, including textile dyeing, electroplating, food processing, and chemical

manufacturing. Grab samples were taken during peak discharge periods over three consecutive days to account for variability in composition. The samples were stored in acid-washed 20 L HDPE containers, maintained at  $4 \pm 2^\circ\text{C}$  during transportation, and analyzed within 24 h to preserve their chemical integrity (Federigi et al. 2024).

The initial analysis of the wastewater involved measuring pH, total dissolved solids (TDS), total suspended solids (TSS), turbidity, chemical oxygen demand (COD), and five-day biological oxygen demand ( $\text{BOD}_5$ ). Six independent wastewater samples were evaluated under raw, DMF, and CAMF conditions. The treatment-level results were expressed as mean  $\pm$  SD calculated from the six independent samples ( $n = 6$ ), following the procedures described in Standard Methods for the Examination of Water and Wastewater (APHA 2017).

Sampling was deliberately carried out during peak discharge periods to assess treatment performance under high-loading conditions typically seen in CETP operations. As a result, the values reported represent typical high-influent operational scenarios rather than long-term seasonal averages. It is important to recognize that industrial influent characteristics can vary seasonally and throughout 24-hour cycles due to production schedules, rainfall, and discharge intermittency. This dataset captures short-term operational variability but does not account for extended seasonal fluctuations. Therefore, the results should be viewed within the context of peak-condition performance, and future research will include longer-term, multi-season monitoring to improve representativeness.

## 2.2. Coagulants and Preparation of Hybrid Coagulant System

A hybrid coagulant system was created by combining two components: aluminum sulfate ( $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ , Merck, India) and *Moringa oleifera* seed powder. Mature *Moringa oleifera* seeds were sourced locally, deshelled, air-dried at room temperature, and ground using a stainless-steel mill. The resulting powder was sieved through a 300  $\mu\text{m}$  mesh and stored in airtight containers to prevent moisture absorption (Ofomi et al. 2025). For stock solution preparation, 10 g of Moringa powder was suspended in 1 L of distilled water, stirred at 150 rpm for 30 min, then allowed to settle for an hour. The clear supernatant was decanted and used as the natural coagulant extract (Desta & Bote 2021).

The coagulant system employed in this study combined aluminum sulfate and *Moringa oleifera* seed extract within a single solution. This approach was designed to leverage complementary coagulation mechanisms: charge neutralization by  $\text{Al}^{3+}$  ions from alum and adsorption-bridging by cationic proteins in Moringa seeds. The hybrid

formulation aimed to improve floc formation, enhance removal efficiency, and diminish membrane fouling potential. Preliminary jar tests determined that the optimal doses were 150  $\text{mg L}^{-1}$  of alum and 250  $\text{mg L}^{-1}$  of Moringa extract, applied simultaneously to the wastewater before filtration (Kane et al. 2016).

### 2.2.1. Coagulant Dose Optimization

Preliminary jar tests using combined alum and *Moringa oleifera* extract were conducted to identify appropriate operating doses prior to membrane experiments. Hybrid combinations of alum at concentrations ranging from 50 to 250  $\text{mg L}^{-1}$  and Moringa extract from 100 to 400  $\text{mg L}^{-1}$  were evaluated. The optimization process considered turbidity reduction, floc formation characteristics such as size and density, settling clarity after 30 min, and residual COD in the supernatant. The optimal doses of 150  $\text{mg L}^{-1}$  alum and 250  $\text{mg L}^{-1}$  Moringa extract achieved effective clarification without producing excessive sludge.

This study evaluated the hybrid formulation as an integrated pretreatment approach; however, the alum-only and Moringa-only systems were not separately optimized within this scope. Therefore, a quantitative assessment of their synergistic interaction was beyond the objectives of this work and is recommended for future research.

## 2.3. Ceramic Membrane Module and Characterization

A commercially available tubular ceramic microfiltration membrane (Membralox®, Pall Corporation, USA) was used in the study. The membrane was composed of  $\alpha$ -alumina and had a nominal pore size of 0.2  $\mu\text{m}$ , an effective surface area of 0.0407  $\text{m}^2$ , and a length of 250 mm. It was installed in a stainless-steel cross-flow module equipped with inlet and outlet pressure gauges to monitor the operating pressure. Filtration was achieved using a variable-speed peristaltic pump (Watson Marlow® 520 series) set to maintain a constant transmembrane pressure (TMP) of 1.0  $\text{kg cm}^{-2}$  at room temperature ( $25 \pm 2^\circ\text{C}$ ), consistent with typical tubular  $\alpha$ -alumina microfiltration setups reported in the literature (Eren & Arslanoğlu 2025).

The physical and surface properties of the ceramic membrane were characterized prior to experimental testing using field-emission scanning electron microscopy (FESEM) combined with energy-dispersive X-ray spectroscopy (EDS) to assess morphology and elemental composition. Crystalline phases were identified via X-ray diffraction (XRD, Rigaku Ultima IV), and surface elemental states were analyzed through X-ray photoelectron spectroscopy (XPS, Thermo Scientific ESCALAB Xi<sup>+</sup>). Total porosity was measured using the Archimedes water-immersion method

in accordance with the protocol described by Hristov et al. (2012).

### 2.3.1. Porosity Measurement

The total porosity ( $\epsilon$ ) of the ceramic membrane was measured using the Archimedes water-immersion technique, as described by Hristov et al. (2012). The dry membrane sample was weighed ( $W_1$ ), then submerged in deionized water under vacuum for 30 min to achieve complete saturation. After removing and surface-drying the sample, its wet weight ( $W_2$ ) was recorded. The saturated membrane was suspended in water, and its apparent weight ( $W_3$ ) was measured with a precision balance. The membrane's porosity was calculated based on these measurements.

$$\epsilon(\%) = \frac{(W_2 - W_1)}{(W_2 - W_3)} \times 100$$

where  $W_1$ ,  $W_2$ , and  $W_3$  represent the dry, wet, and suspended weights of the membrane, respectively.

All measurements were conducted in triplicate, and mean values  $\pm$  standard deviation were reported. The obtained porosity ( $43 \pm 2\%$ ) was consistent with values typical of mesoporous  $\alpha$ -alumina supports.

## 2.4. Filtration Experiments

### 2.4.1. Direct Membrane Filtration (DMF)

In the direct filtration mode, untreated wastewater was fed to the membrane module without pretreatment. Each run was carried out for 60 min under a fixed TMP of  $1.0 \text{ kg cm}^{-2}$ . Permeate was collected at 10-min intervals using a digital balance connected to a data logger to record cumulative mass. The instantaneous permeate flux ( $J$ ,  $\text{L m}^{-2} \text{ h}^{-1}$ ) was calculated as:

$$J = \frac{V}{At}$$

where  $V$  is the volume of permeate collected (L),  $A$  is the effective membrane area ( $\text{m}^2$ ), and  $t$  is filtration time (h) (Sanchis-Perucho et al. 2023).

### 2.4.2. Coagulant-Assisted Membrane Filtration (CAMF)

In the coagulant-assisted mode, a hybrid coagulant system consisting of alum and Moringa was directly added to the wastewater. The mixture was rapidly stirred at 200 rpm for 2 min, then gently mixed at 40 rpm for 20 min. Following this, it was allowed to settle for 30 min. The clarified supernatant was subsequently filtered through a membrane under the same conditions used for DMF (Singh et al. 2023). The combined action of aluminum sulfate and Moringa extract was anticipated to decrease colloidal particles, increase floc

density, and prevent pore blockage on the ceramic surface (Katalo et al. 2018).

## 2.5. Membrane Cleaning and Performance Recovery

After each filtration cycle, the membrane module was rinsed with deionized water at a pressure of  $1.5 \text{ kg cm}^{-2}$  for 10 min to eliminate reversible fouling layers. Weekly chemical cleaning involved using  $0.1 \text{ M NaOH}$  at  $40^\circ \text{C}$  for 30 min, followed by rinsing until the effluent reached a neutral pH. The clean-water flux (CWF) was measured before and after cleaning to assess fouling reversibility and membrane recovery efficiency (Ezugbe & Rathilal 2020).

## 2.6. Analytical Determinations and Data Analysis

All influent and permeate samples were analyzed for pH, TSS, TDS, turbidity, COD, and BOD<sub>5</sub> following APHA (2017) procedures. Pollutant removal efficiency was then calculated as

$$\% \text{ Removal} = \frac{C_i - C_f}{C_i} \times 100,$$

where  $C_i$  and  $C_f$  are the influent and effluent concentrations, respectively. Six independent wastewater samples were analyzed under raw, DMF, and CAMF conditions, and the results were expressed as mean  $\pm$  SD. Statistical analysis was conducted using OriginPro 2022b. Data normality was assessed prior to analysis. For normally distributed data, one-way ANOVA was applied. Non-normally distributed data were analyzed using the Kruskal–Wallis test. Flux was analyzed using an independent samples t-test. Statistical significance was accepted at  $p < 0.05$ .

# 3. RESULTS AND DISCUSSION

## 3.1. Membrane Characterization

The microstructural and compositional features of the alumina ceramic microfiltration membrane were analyzed to establish a relationship between material characteristics and filtration efficiency. FESEM imaging (Fig. 1(a)) displayed a consistent pore structure typical of sintered  $\alpha$ - $\text{Al}_2\text{O}_3$  supports. EDS elemental mapping (Fig. 1(b)) verified the dominance of aluminum and oxygen, with minor silicon traces originating from bonding materials. Post-filtration surfaces exhibited distinct fouling morphologies:

Following filtration experiments, the membrane surfaces exhibited distinct fouling morphologies, consisting of deposited particulates and surface films that varied depending on feed composition.

X-ray diffraction (XRD) analysis (Fig. 2) exhibited sharp peaks corresponding to  $\alpha$ - $\text{Al}_2\text{O}_3$  at  $2\theta = 25.5^\circ$ ,  $35.1^\circ$ ,

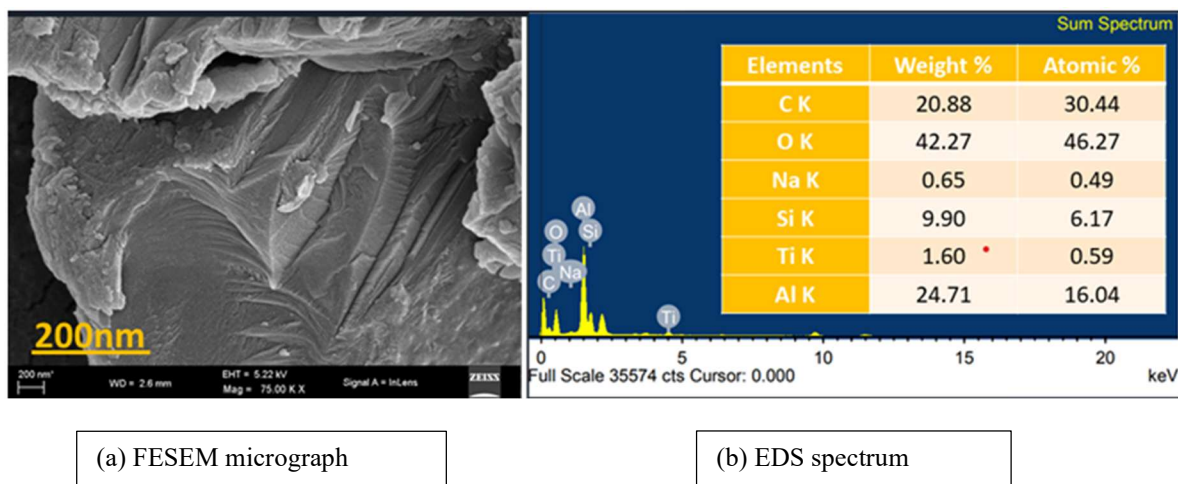


Fig. 1: (a) FESEM micrograph of the  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> support surface showing a uniform, interconnected pore structure at the nanoscale (scale bar: 200 nm); (b) EDS spectrum and quantitative elemental composition confirming the predominance of Al and O consistent with Al<sub>2</sub>O<sub>3</sub>, with minor Si, Na, Ti, and C originating from binder residues and surface contaminants.

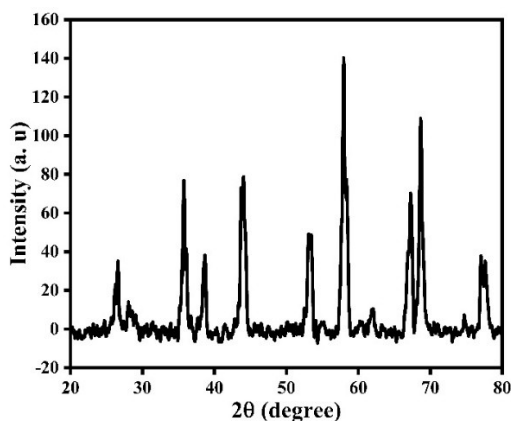


Fig. 2: XRD pattern of the commercial  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> membrane showing characteristic diffraction peaks of the  $\alpha$ -alumina phase.

37.8°, 43.4°, 52.6°, 57.5°, 66.5°, and 76.9°, confirming the crystalline integrity of the alumina phase (JCPDS No. 46-1212). No secondary impurity phases were detected.

### 3.1.1. XPS Analysis

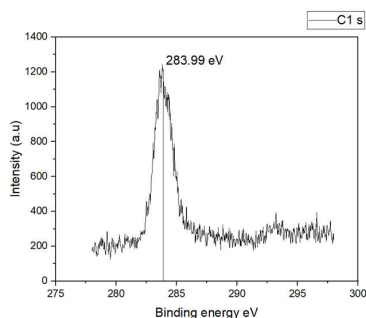
X-ray photoelectron spectroscopy (XPS) was used to determine the surface elemental composition and oxidation states of the ceramic membrane (Fig. 3(a–f)). The survey spectrum confirmed the presence of Al, O, C, Si, Na, and Ti, in agreement with the EDS results.

The high-resolution C 1s spectrum (Fig. 3(a)) exhibits a dominant peak centered at 283.99 eV, characteristic of surface carbon species typically attributed to atmospheric hydrocarbon adsorption. The absence of significant higher-binding-energy components indicates negligible carbonate or oxygenated carbon contamination.

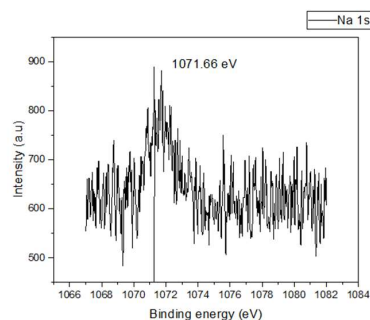
The Na 1s spectrum (Fig. 3(b)) shows a peak at 1071.66 eV, consistent with sodium present in its ionic form (Na<sup>+</sup>). The relatively weak intensity suggests trace-level sodium, likely originating from precursor materials or processing residues rather than from a distinct sodium-rich phase.

The Si 2p peak (Fig. 3(c)), located at 102.56 eV, corresponds to Si<sup>4+</sup> species in a silicate environment, confirming the presence of silica-based binders or minor secondary phases within the membrane structure.

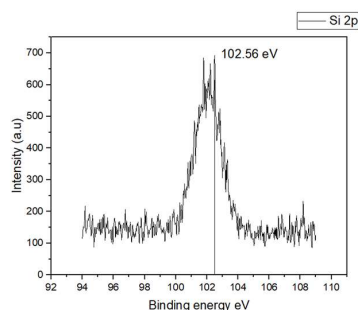
The Ti 2p region (Fig. 3(d)) displays a Ti 2p<sub>3/2</sub> peak centered at 457.98 eV, indicative of Ti<sup>4+</sup> in TiO<sub>2</sub>. No lower-binding-energy contributions associated with Ti<sup>3+</sup> or metallic Ti were observed, suggesting that titanium is present predominantly in its fully oxidized state.



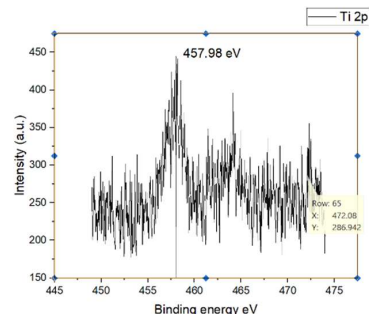
(a) C 1s XPS spectrum with peak at 283.99



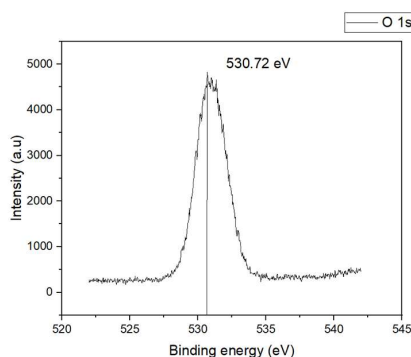
(b) Na 1s XPS spectrum with peak at



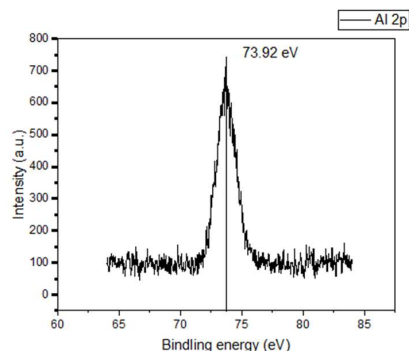
(c) Si 2p XPS spectrum with peak at 102.56 eV.



(d) Ti 2p XPS spectrum with peak at 457.98 eV.



(e) O 1s XPS spectrum with peak at 530.72 eV.



(f) Al 2p XPS spectrum with peak at 73.92 eV.

Fig. 3: High-resolution XPS spectra of the ceramic membrane: (a) C 1s, (b) Na 1s, (c) Si 2p, (d) Ti 2p, (e) O 1s, and (f) Al 2p.

The O 1s spectrum (Fig. 3(e)) presents a strong, symmetrical peak at 530.72 eV, characteristic of lattice oxygen ( $O^{2-}$ ) in metal oxides. The narrow peak profile indicates minimal surface hydroxylation or adsorbed oxygen species.

The Al 2p spectrum (Fig. 3(f)) shows a sharp peak at 73.92 eV, consistent with  $Al^{3+}$  in  $\alpha-Al_2O_3$ . This binding energy confirms that aluminum exists predominantly in its stable oxide form and constitutes the primary structural framework of the membrane.

Table 1: Summary of pore characteristics and surface area measurements of the ceramic microfiltration membrane.

| Parameter                                     | Method                          | Result | Unit                            |
|-----------------------------------------------|---------------------------------|--------|---------------------------------|
| BET surface area                              | N <sub>2</sub> adsorption (BET) | 90.7   | m <sup>2</sup> g <sup>-1</sup>  |
| Langmuir surface area                         | Langmuir model                  | 97.9   | m <sup>2</sup> g <sup>-1</sup>  |
| BJH cumulative surface area (pore > 2 nm)     | BJH model                       | 103.0  | m <sup>2</sup> g <sup>-1</sup>  |
| Total pore volume (P/P <sub>0</sub> = 0.9987) | N <sub>2</sub> adsorption       | 0.9469 | cm <sup>3</sup> g <sup>-1</sup> |
| Average pore diameter                         | BET                             | 21.0   | nm                              |
| Micropore surface area                        | DR model                        | 139.8  | m <sup>2</sup> g <sup>-1</sup>  |
| Porosity (Archimedes method)                  | Water immersion                 | 43 ± 2 | %                               |

Collectively, the high-resolution XPS spectra confirm that the membrane surface is dominated by stable oxide phases (Al–O, Ti–O, and Si–O bonding), with only trace levels of residual sodium and surface carbon. These results further validate the chemical integrity and compositional stability of the  $\alpha$ -alumina ceramic matrix.

BET analysis yielded a specific surface area of 90.7 m<sup>2</sup> g<sup>-1</sup>, a total pore volume of 0.9469 cm<sup>3</sup> g<sup>-1</sup>, and an average pore diameter of ~21 nm. The porosity, determined using the Archimedes method, was 43 ± 2%, which is characteristic of mesoporous alumina ceramics. The corresponding data are summarized in Table 1.

These structural properties, confirmed by complementary XRD and XPS analysis, underpin the observed filtration efficiency and fouling reversibility, as discussed in Sections 3.2 and 3.3.

### 3.2. Physicochemical Characteristics of Raw Wastewater

The composite industrial effluent collected from the CETP influent exhibited highly variable physicochemical characteristics, as expected for mixed discharges from textile, electroplating, and food-processing units. The raw wastewater was slightly acidic, with a pH ranging from 6.0 to 6.5. The average COD (4688 ± 280 mg L<sup>-1</sup>) and BOD<sub>5</sub> (1523 ± 82 mg L<sup>-1</sup>) concentrations confirmed a high organic load, typical of industrial clusters. Elevated TSS (450 ± 28 mg L<sup>-1</sup>) and TDS (6875 ± 220 mg L<sup>-1</sup>) levels reflected substantial suspended and dissolved solids, while a turbidity of approximately 215 NTU indicated intense coloration and colloidal instability. These results underline the need for pretreatment to improve effluent compatibility with biological processes (APHA 2017).

### 3.3. Direct Membrane Filtration (DMF) Performance

Direct ceramic microfiltration of untreated wastewater produced a moderate improvement in water clarity and solids removal but achieved limited organic reduction. The mean turbidity declined from 215 NTU to 59 NTU

(approximately 72%), while TSS decreased from 450 ± 28 mg L<sup>-1</sup> to 28 ± 2 mg L<sup>-1</sup>, corresponding to approximately 93.8% removal. COD and BOD<sub>5</sub> removal under DMF were approximately 22.7% and 25.0%, respectively, consistent with the membrane's ability to retain suspended particles while allowing a substantial fraction of dissolved organic matter to pass through. The pH increased slightly to 6.5 ± 0.2, likely due to the partial removal of acidic particulates.

The steady-state flux recorded for DMF averaged 45 ± 2 L m<sup>-2</sup> h<sup>-1</sup> under a constant transmembrane pressure of 1.0 kg cm<sup>-2</sup>. A rapid initial decline in flux occurred within the first 15 min, after which the flux stabilized, indicating cake-layer formation as the dominant fouling mechanism. Similar trends have been observed in ceramic membranes treating high-strength industrial effluents (Kapila et al. 2025). Although flux recovery after hydraulic cleaning reached approximately 87%, significant reversible fouling persisted, highlighting the limitations of DMF for treating untreated complex wastewater.

The moderate COD removal observed in DMF is consistent with the intrinsic pore-size limitation of microfiltration membranes (~0.2 µm), which primarily retain suspended and colloidal matter while allowing dissolved organics to pass through. This explains the limited reduction in dissolved organic load despite the effective removal of turbidity.

### 3.4. Coagulant-Assisted Membrane Filtration (CAMF) Using Combined Alum and *Moringa oleifera*

Pretreatment of the wastewater using a combined coagulant mixture of aluminum sulfate and *Moringa oleifera* seed extract produced a distinct improvement in both water quality and filtration performance. The hybrid coagulant promoted the formation of large, dense flocs through complementary mechanisms: charge neutralization by Al<sup>3+</sup> ions and adsorption–bridging by cationic *Moringa* proteins (Kane et al. 2016).

Following CAMF, turbidity was reduced to 17 ± 2 NTU (approximately 92% removal), while TSS declined from 450 ± 28 mg L<sup>-1</sup> to 9 ± 1 mg L<sup>-1</sup>, corresponding to approximately

98.0% removal. COD and BOD<sub>5</sub> decreased markedly to  $855 \pm 55 \text{ mg L}^{-1}$  and  $305 \pm 18 \text{ mg L}^{-1}$ , respectively, corresponding to average reductions of 81.8% and 80.0% compared with the raw influent. The effluent pH stabilized at 6.8–7.0, a range conducive to downstream biological oxidation (Zielińska & Galik 2017).

The average steady-state flux during CAMF increased to approximately  $71 \text{ L m}^{-2} \text{ h}^{-1}$ , around 55% higher than that of DMF, demonstrating effective fouling mitigation. This improvement resulted from the reduced colloidal loading and the formation of compact, settleable flocs that limited pore blockage (Aragaw & Bogale 2023).

The improved flux behavior under CAMF conditions can be attributed to the modification of the feed colloidal matrix prior to membrane contact. Alum promotes charge neutralization of negatively charged particles, while bioactive proteins in *Moringa* extract facilitate adsorption–bridging. The resulting flocs are larger and less compressible,

producing a more permeable cake structure and reducing pore-blocking tendencies.

### 3.5. Flux Behavior

The variation in permeate flux with time during microfiltration is presented in Fig. 4. Both direct membrane filtration (DMF) and coagulant-assisted microfiltration (CAMF) exhibited a sharp initial decline in flux within the first 10 min, which was attributed to rapid pore blocking and cake-layer formation by suspended solids and colloidal matter. Thereafter, the flux reached a quasi-steady state due to the establishment of a dynamic filtration layer.

The CAMF system consistently maintained a higher permeate flux ( $\sim 71 \text{ L m}^{-2} \text{ h}^{-1}$ ) than DMF ( $\sim 45 \text{ L m}^{-2} \text{ h}^{-1}$ ) throughout the 60-min filtration run, indicating improved fouling control. This enhancement is attributed to the pre-aggregation of colloidal and organic matter by the natural coagulant (*Moringa oleifera*) prior to membrane contact,

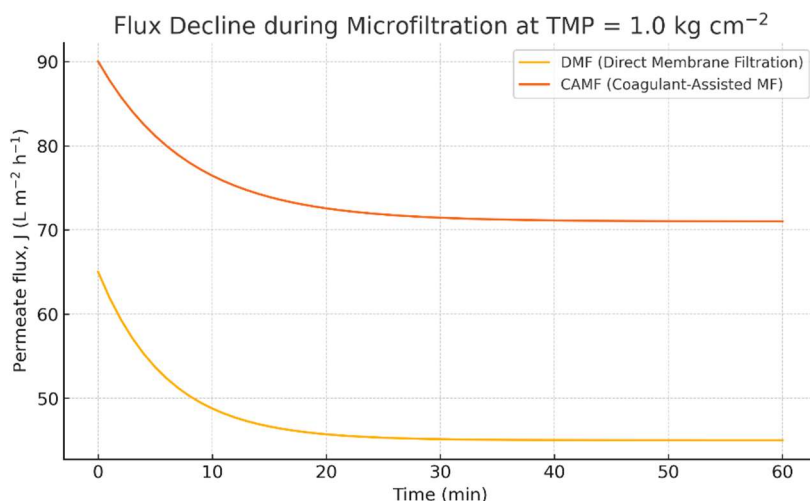


Fig. 4: Comparison of flux decline profiles for DMF and CAMF at  $1.0 \text{ kg cm}^{-2}$ , highlighting slower flux decay and higher steady-state flux under CAMF conditions.

Table 2 summarizes the mean  $\pm$  SD values calculated from six independent wastewater samples ( $n = 6$ ) evaluated under raw, DMF, and CAMF conditions.

| Parameter                                 | Raw effluent (mean $\pm$ SD) | DMF (mean $\pm$ SD) | CAMF (mean $\pm$ SD) | Statistic           | p-value     |
|-------------------------------------------|------------------------------|---------------------|----------------------|---------------------|-------------|
| pH                                        | $6.25 \pm 0.18$              | $6.46 \pm 0.21$     | $6.80 \pm 0.20$      | 13.798 <sup>f</sup> | $p < 0.001$ |
| COD ( $\text{mg L}^{-1}$ )                | $4688 \pm 280$               | $3625 \pm 180$      | $855 \pm 55$         | 15.158 <sup>h</sup> | 0.001       |
| BOD <sub>5</sub> ( $\text{mg L}^{-1}$ )   | $1523 \pm 82$                | $1143 \pm 46$       | $305 \pm 18$         | 15.174 <sup>h</sup> | 0.001       |
| TSS ( $\text{mg L}^{-1}$ )                | $450 \pm 28$                 | $28 \pm 2$          | $9 \pm 1$            | 15.237 <sup>h</sup> | $p < 0.001$ |
| TDS ( $\text{mg L}^{-1}$ )                | $6875 \pm 220$               | $6560 \pm 160$      | $6133 \pm 130$       | 12.904 <sup>h</sup> | 0.002       |
| Turbidity (NTU)                           | $215 \pm 8$                  | $59 \pm 3$          | $17 \pm 2$           | 15.205 <sup>h</sup> | $p < 0.001$ |
| Flux ( $\text{L m}^{-2} \text{ h}^{-1}$ ) | –                            | $45 \pm 2$          | $71 \pm 3$           | 39.51 <sup>t</sup>  | $p < 0.001$ |

Note: <sup>f</sup> F value from one-way ANOVA; <sup>h</sup> H value from the Kruskal–Wallis test; <sup>t</sup> t value from the independent-samples t-test.

which reduced direct pore deposition and the compressibility of the cake layer.

The lower rate of flux decline in CAMF demonstrates the synergistic effect of coagulation and membrane separation, as the coagulant modifies the particle size distribution and surface charge, resulting in a more permeable cake structure. This behavior is consistent with previous reports on hybrid coagulation–membrane systems for industrial wastewater treatment.

The observed flux stabilization also indicates the partial reversibility of fouling, suggesting that periodic backwashing or hydraulic cleaning can effectively restore flux without significant deterioration in membrane performance.

### 3.6. Comparative Performance and Statistical Evaluation

Table 2: Physicochemical characteristics of raw effluent and effluent treated with DMF and CAMF membranes. Values are presented as mean  $\pm$  SD ( $n = 6$  independent wastewater samples). Data normality was assessed prior to analysis. pH was analyzed using one-way ANOVA. COD, BOD<sub>5</sub>, TSS, TDS, and turbidity were analyzed using the Kruskal–Wallis test. Flux was analyzed using an independent samples t-test.

The data indicate that CAMF substantially improved organic matter removal compared with DMF, while also providing a further reduction in TSS and maintaining a significantly higher flux. Statistical analysis confirmed significant differences among the raw effluent, DMF, and CAMF samples for COD, BOD<sub>5</sub>, TSS, TDS, and turbidity ( $p < 0.05$ ). The marginal change in TDS indicates that microfiltration primarily removes suspended and colloidal particles rather than dissolved ions (Uman et al. 2024).

The improvement in flux resulted from reduced pore constriction and lower cake resistance, verifying the advantage of pre-coagulation in membrane operations. Similar flux enhancement has been reported with the combined use of inorganic and bio-coagulants, which was attributed to the formation of more permeable floc structures (Singh et al. 2023). Comparable trends have also been reported in other hybrid coagulation–membrane studies, where pre-aggregation of colloids reduced cake resistance and improved hydraulic stability. The present findings are consistent with these observations and support the potential suitability of hybrid pretreatment for treating variable industrial effluents.

### 3.7. Membrane Fouling and Cleaning Behavior

After each run, the membrane was hydraulically rinsed and chemically cleaned. Clean-water flux recovery reached 95% after CAMF, compared with 87% after DMF, confirming

that hybrid coagulant pretreatment produced a less compact fouling layer and minimized irreversible resistance (Katalo et al. 2018). The hybrid system employed 150 mg L<sup>-1</sup> of alum in combination with 250 mg L<sup>-1</sup> of Moringa extract; however, because an independently optimized alum-only system was not evaluated, quantitative alum savings relative to conventional alum-only treatment could not be established in the present study. Direct comparison of optimized alum-only, Moringa-only, and hybrid coagulation systems would be required to quantify potential coagulant savings and establish any synergistic effect.

Overall, these findings validate the effectiveness of integrating hybrid coagulation with ceramic microfiltration for CETP influent pretreatment. The process provides higher organic and solids removal, improved flux stability, and favorable pH conditions, ensuring compatibility with subsequent biological treatment stages.

From an operational perspective, the stabilization of pH toward neutrality and the reduction in suspended solids are particularly beneficial for downstream biological units in CETPs. Lower shock loading and improved influent consistency can reduce biomass inhibition and enhance process reliability under fluctuating industrial discharge conditions.

### 3.8. Fouling Mechanism Interpretation

Although detailed resistance quantification was not performed, the fouling behavior can be interpreted using the classical resistance-in-series concept, in which the total hydraulic resistance is considered the sum of the intrinsic membrane resistance and fouling resistance. In the present study, fouling development was inferred from the flux decline profiles and flux recovery following cleaning.

The sharp initial decrease in permeate flux during the early stage of filtration suggests rapid surface deposition of suspended solids and colloidal matter. The subsequent stabilization of flux indicates the formation of a dynamic cake layer that governed the steady-state resistance rather than continuous internal pore constriction.

Flux recovery after hydraulic rinsing demonstrates that a significant proportion of the fouling was reversible and attributable to loosely attached cake deposits. The higher recovery observed under coagulant-assisted conditions suggests that pre-aggregation of particles reduced compact pore blocking and limited the development of irreversible resistance.

Although explicit resistance partitioning into membrane resistance, reversible fouling resistance, and irreversible fouling resistance would require systematic clean-water flux measurements before and after each filtration cycle, the

observed filtration behavior supports cake-dominated fouling under the investigated operating conditions.

Future work involving resistance modeling and pore-blocking kinetics would further strengthen the mechanistic understanding of the process at the pilot scale.

### 3.9. Study Limitations

Although the hybrid system demonstrated significant improvements in the removal of organic matter and suspended solids, dissolved solids removal remained limited, reflecting the inherent constraints of microfiltration. Long-term fouling behavior and a detailed techno-economic evaluation were beyond the scope of the present study and warrant further investigation before large-scale implementation.

### 3.10. Practical and Scale-Up Considerations

For integration into existing CETP infrastructure, operational and sludge management aspects must be considered. The hybrid coagulation step generates sludge consisting of aluminum hydroxide flocs combined with aggregated suspended solids and organic matter bridged by *Moringa*-derived biopolymers. Although the sludge yield was not quantified in the present laboratory-scale study, the applied alum dose ( $150 \text{ mg L}^{-1}$ ) falls within the moderate range typically used for industrial coagulation and is lower than the doses commonly required for highly turbid industrial effluents. Partial use of the natural bio-coagulant alongside alum may have the potential to reduce reliance on inorganic coagulant dosing; however, the resulting sludge quantity and composition were not quantified in the present study.

The generated sludge would require conventional thickening and dewatering before disposal or potential co-processing, consistent with standard CETP sludge management practices. The organic fraction introduced by *Moringa* extract is biodegradable and may not significantly increase the hazardous classification of the sludge, although detailed sludge characterization remains necessary for full-scale implementation.

From an operational perspective, the microfiltration system operated at  $1 \text{ kg cm}^{-2}$  (approximately 1 bar), corresponding to low-pressure membrane operation. Because electrical energy consumption was not directly measured in the present laboratory-scale experiments, a system-specific energy consumption could not be established. For preliminary scale-up assessment, a literature-based value of approximately  $0.2 \text{ kWh m}^{-3}$  may be used as a representative MF/UF energy-consumption estimate (Tow et al. 2021). This value is adopted only as an indicative literature value and does not represent experimentally measured energy consumption for the present ceramic membrane system.

The actual energy demand will depend on pump efficiency, cross-flow velocity, transmembrane pressure, membrane resistance, and recovery; therefore, pilot-scale measurements are required for system-specific assessment.

Under the optimized conditions, chemical consumption corresponded to  $150 \text{ mg}$  of alum and  $250 \text{ mg}$  of *Moringa* extract per L of influent. For a representative CETP treating  $1,000 \text{ m}^3 \text{ day}^{-1}$ , this would translate to approximately  $150 \text{ kg day}^{-1}$  of alum and  $250 \text{ kg day}^{-1}$  of *Moringa* extract, subject to influent variability and further optimization.

In terms of footprint, tubular ceramic membrane modules offer a more compact installation than large sedimentation tanks, making retrofitting feasible in space-constrained industrial clusters. However, a comprehensive techno-economic assessment and long-term operational trials are necessary to validate the cost-effectiveness of the process at full scale.

## 4. CONCLUSIONS

This study assessed the feasibility of combining hybrid coagulation using aluminum sulfate with *Moringa oleifera* extract and ceramic microfiltration as a pretreatment method for mixed industrial wastewater in a Common Effluent Treatment Plant (CETP). Under the tested conditions, the integrated approach yielded superior removal of suspended solids, turbidity, and organic load compared to direct membrane filtration alone. The improved steady-state flux and higher flux recovery after cleaning suggest a lower fouling tendency when coagulation is performed before membrane filtration.

The findings indicate that hybrid coagulation–microfiltration effectively stabilizes high-strength composite industrial influent during peak loading conditions. However, these results are derived from short-term laboratory experiments using a single membrane setup and limited sampling over time. Consequently, the outcomes should be viewed as a proof-of-concept rather than indicative of full-scale performance.

While the approach shows potential for improving influent consistency prior to biological treatment, further work is required to assess long-term fouling dynamics, sludge generation characteristics, seasonal variability, and techno-economic feasibility under continuous operation.

Overall, the study offers initial evidence that combining hybrid coagulation with ceramic microfiltration is a promising pretreatment method for complex industrial wastewater, justifying further pilot-scale testing.

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