

## A study of inline chemical coagulation/precipitation-ceramic microfiltration and nanofiltration for reverse osmosis concentrate minimization and reuse in the textile industry

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### ABSTRACT

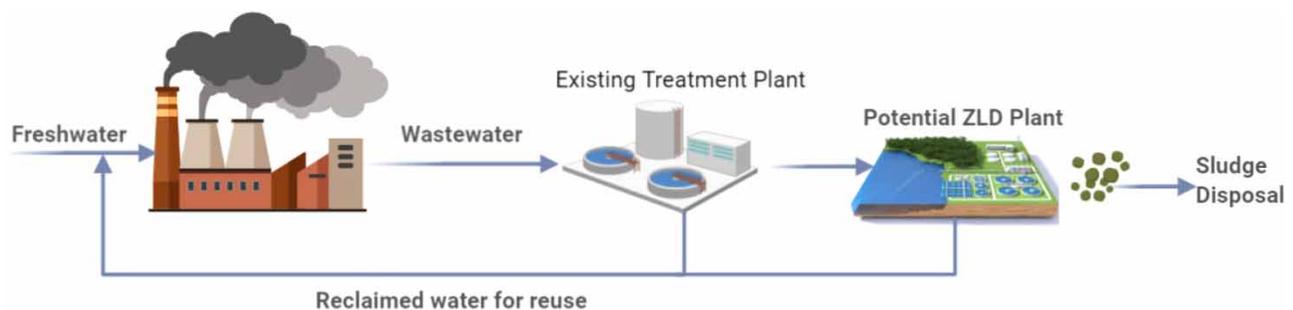
Reverse osmosis concentrate (ROC) is one of the major drawbacks in membrane treatment technologies specifically due to the scale-forming ions. It is important to remove these ions from ROC to enhance total water recovery and reuse in the textile industry that is the largest water-consumer and polluter industry. In this work, coagulation/high pH precipitation (CP) integrated with ceramic microfiltration (CMF) was studied as a pre-treatment method followed by nanofiltration (NF) to increase the efficiency of water recovery. To prevent organic fouling, ferric chloride ( $\text{FeCl}_3$ ) was applied at a concentration of 3 mM, and ceramic membranes were used for the removal of non-precipitating crystals and/or suspended solids (at high pH) before the NF processes. The CP-CMF method successfully removed calcium ( $\text{Ca}^{2+}$ ), magnesium ( $\text{Mg}^{2+}$ ), silica ( $\text{SiO}_2$ ), and TOC up to 97, 83, 92, and 87% respectively, which resulted in higher performance of the NF process. Moreover, this method provided higher flux at lower pressure that ultimately increased overall water recovery of the NF process to achieve near-zero liquid discharge (n-ZLD). A cost-benefit estimation showed that a high-quality effluent (COD < 5 mg/L; conductivity  $700 < \mu\text{S}/\text{cm}$ ; negligible residual color) can be generated and recycled in the textile industry at an economical cost (approximately 0.97 USD/m<sup>3</sup>). Therefore, ROC minimization and water recovery can help to achieve n-ZLD using the CP-CMF/NF method.

**Key words:** cost estimation, membrane filtration, near-zero liquid discharge, reverse osmosis concentrate, silica removal, textile wastewater

### HIGHLIGHTS

- Studied reverse osmosis concentrate (ROC), an emerging hotspot of pollution.
- Explored the removal of scaling precursors and concentrations of organics.
- Investigated minimization of ROC by increasing the efficiency of membrane processes.
- Discussed cost-benefit estimation to find the economic feasibility of the adopted method.

### GRAPHICAL ABSTRACT



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## LIST OF ABBREVIATIONS

Ca(OH) <sub>2</sub>	Calcium hydroxide
Ca <sup>2+</sup>	Calcium
CMF	Ceramic microfiltration
COD	Chemical oxygen demand
EC	Electrical conductivity
FeCl <sub>3</sub>	Ferric chloride
J	Wastewater flux
J/J <sub>0</sub>	Flux decline
J <sub>0</sub>	Wastewater initial flux
kDa	Kilodalton
MF	Microfiltration
Mg <sup>2+</sup>	Magnesium
MgO	Magnesium oxide
MWCO	Molecular weight cut-off
NaOH	Sodium Hydroxide
NF	Nanofiltration
Pt–Co	Platinum–Cobalt
R	Resistance
RO	Reverse osmosis
ROC	Reverse osmosis concentrate
Sal%	Salinity (%)
SiO <sub>2</sub>	Silica
TOC	Total organic carbon
UF	Ultrafiltration

## 1. INTRODUCTION

Due to rapid growth in population, urbanization, and industrialization, the reuse of industrial wastewater has become a trend for sustainable water management strategies to reduce freshwater consumption and minimize contamination of receiving water bodies (Yaquub & Lee 2019). The textile industry is one of the largest water-consumer industries, the reclamation of textile wastewater using a membrane-based reverse osmosis (RO) technology is widespread these days (Sahinkaya *et al.* 2018). This technology for the recovery of water and valuable salts especially in intensive water-consuming sectors like the textile industry has incited deep interest because of permeate quality (Joo & Tansel 2015), energy consumption, and safe operation (Yu *et al.* 2013). In addition, the best available technique reference documents (BREFs) emphasized an application of membrane technologies for effective water and salt recovery in the textile industry (Ozturk *et al.* 2016). However, one of the main limitations in RO application is the disposal of the RO concentrate (ROC) because it has a much higher concentration of contaminants as compared to the feed water (Zhao *et al.* 2019). The ROC ranges 15–50% of feed water which must be disposed of or treated for reuse (Warsinger *et al.* 2018). If untreated or improperly managed ROC is discharged to water bodies and it can cause adverse effects such as eutrophication, high salinity (Joo & Tansel 2015), and organic pollution (Tran *et al.* 2012) in the aquatic environment. Additionally, stringent environmental regulations are forcing the textile sector to reuse the ROC after further treatment to achieve zero liquid discharge (ZLD).

Membrane fouling is one of the most common issues, and it refers to the deposition or adsorption of contaminants on the membrane surface (Shon *et al.* 2002). Organic contaminants with particle sizes larger than the pores of the membrane are easily adsorbed on the membrane's surface during filtration (Elimelech & Wiesner 2002). This can happen due to the following reasons: (1) colloidal fouling; (2) organic fouling due to the deposition of organic substances; and (3) scaling, which is known as the formation of mineral deposits precipitating from the feed stream onto the membrane surface results in a loss of permeate flux through the membrane (Duranceau 2001). Due to these reasons, minimizing the scale-forming ions in ROC for further treatments is a significant issue. Since the presence of divalent cations is crucial in the membrane fouling process, it is critical to understand their function and mechanism. Calcium and magnesium ions, which are the most common and abundant cations in water settings, are the most representative divalent inorganic ions (Meng *et al.* 2020). In a recent study, the effect of Mg<sup>2+</sup> on polysaccharide fouling during ultrafiltration was investigated, and it was discovered that gel layer formation was the dominant fouling mechanism in the presence of Mg<sup>2+</sup> (Wang *et al.* 2020). Therefore, pre-treatment before the membrane process is an attractive option to reduce the risk of fouling that ultimately increases treatment efficiency

(Semblante *et al.* 2018). The primary target for pre-treatment of ROC is the removal of divalent ions (Sahinkaya *et al.* 2018) and organic matter (Umar *et al.* 2016) as studied by the addition of chemicals (Subramani & Jacangelo 2014). Coagulation has shown a great effect on organic matter removal but not sufficient for dealing with the high concentrations of scale-forming ions in ROC (Ho *et al.* 2015). The use of single treatment technology to achieve ZLD/near-ZLD is not possible for ROC treatment and thus a combination of technologies is essential (Subramani & Jacangelo 2014). The selection of the best pre-treatment method for further treatment of ROC to increase water recovery depends on the process requirements (Pérez-González *et al.* 2012). For instance, treatment of ROC through membrane filtration requires the removal of organics and scale precursors to prevent scaling and fouling (Semblante *et al.* 2018). As a result, a combination of coagulation with chemical precipitation could be useful for creating a synergetic pre-treatment (Ordóñez *et al.* 2012). By comparison, ceramic MF/UF membranes have the potential for pre-treatment when combined with oxidation processes to remove pollutants and prevent membrane fouling (Gray *et al.* 2015).

Some researchers have widely studied state-of-the-art technologies for RO concentrate minimization. Recent studies have investigated using a seeded precipitation stage that has been integrated into the ceramic ultrafiltration process for the removal of precipitated  $\text{Ca}^{2+}$  to prevent membrane fouling and increase water recovery (Sanciolo *et al.* 2012). Another simultaneous treatment hybrid system, consisting of coagulation with  $\text{FeCl}_3$  + photocatalysis (UVC/ $\text{TiO}_2$ ) was used and this system achieved 95% organic matter removal from the ROC within 6 hours (Zhou *et al.* 2011). Using a pellet reactor achieved 85%  $\text{Ca}^{2+}$  removal at a pH of 11.5, this removal was increased up to 95% when the reactor was combined with electro dialysis (ED) (Tran *et al.* 2012). The coagulation process in combination with the UV/ $\text{H}_2\text{O}_2$  system achieved 17–27% additional DOC removal and suggested  $\text{FeCl}_3$  as the most efficient coagulant (Umar *et al.* 2016). Some researchers achieved 80%  $\text{SiO}_2$  removal from brackish water ROC using electro-coagulation under a current intensity of 0.5 (A) for 30 minutes of hydraulic retention time (Den & Wang 2008) and a seeded precipitation process was applied with a two-stage RO system, which presented 98% water recovery (Gabelich *et al.* 2007).

Generally, ROC studies have been carried out with groundwater, desalination, and municipal wastewater reclamation. Due to the presence of high color, organic matter, toxic compounds, and salt concentrations, textile ROC is quite difficult to treat (Holkar *et al.* 2016). The novelty of the research is the application of CMF and its combination with CP for the removal of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{SiO}_2$  from textile ROC. This work aims to (1) investigate the CMF pre-treatment to remove the precipitated scaling ions, (2) explore the efficiency of CP and CMF integrated system from textile ROC streams, (3) prevent membrane fouling using CP-CMF pre-treatment, and (4) conduct cost-benefit analysis for economic feasibility to scale-up the system.

## 2. MATERIALS AND METHODS

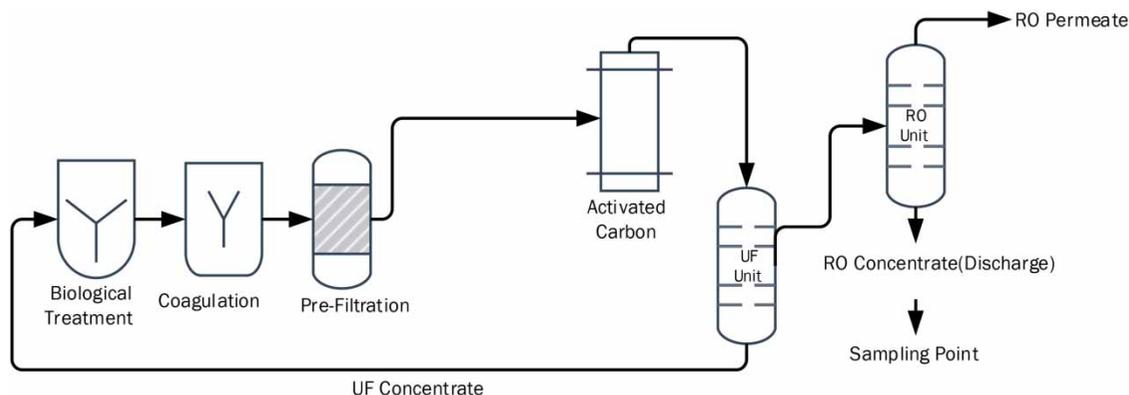
### 2.1. Sampling site and wastewater characterization

Characterization and filtration experiments were conducted with real ROC samples that were collected from the household linen processing textile industry. This industry is located in Tekirdag, Turkey with a capacity of 5,000  $\text{m}^3/\text{d}$  wastewater treatment. The treatment system consists of mechanical-physical treatment followed by biological treatment (activated sludge process). At this plant, biologically treated wastewater runs through the coagulation-pre-filtration unit and activated carbon adsorption unit, before undergoing UF and RO membrane processes. RO permeate is used for cooling tower water requirements. The existing wastewater treatment system scheme selected for this study is shown in Figure 1.

Ten samples of the ROC stream were collected and tested for COD, TOC, pH, EC, color, total hardness,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{SiO}_2$  analysis. Measured characteristics were in the range as reported in the literature for pH, COD, conductivity,  $\text{Na}^+$ ,  $\text{SO}_4^{2-}$  (Sahinkaya *et al.* 2018), and TDS, TP, and  $\text{Cl}^-$  (Dhodapkar *et al.* 2007). Differences were observed in silica and  $\text{Mg}^{2+}$  concentrations, in the present study, silica concentration was higher and  $\text{Mg}^{2+}$  concentration was lower than similar effluent average values. The characteristics of textile ROC in comparison with literature studies are presented in Table 1.

### 2.2. Experimental setup

All the chemicals used in the study were of analytical grade and purchased from Merck, Germany ( $\text{FeCl}_3$ ,  $\text{MgO}$ ,  $\text{Ca}(\text{OH})_2$ ), Sigma Aldrich ( $\text{NaOH}$ ), and Carlo Erba ( $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ ). The stock solution of  $\text{FeCl}_3$  and  $\text{NaOH}$  was prepared with ultrapure water (18.18  $\text{m}\Omega\text{-cm}$ ). In this study, we used tubular type ceramic membranes with three different molecular weight cut-off (MWCO) (0.2  $\mu\text{m}$ , 15 kDa, 300 kDa), which were procured from TAMI Industries (Nyons, France) and polymeric NF membranes (NF270 and NF90) from Dow Filmtec™. The specifications of membranes are presented in Table 2.



**Figure 1** | Studied textile industry existing wastewater treatment plant scheme.

**Table 1** | Textile ROC characterization in the current study and literature values

Parameter	Units	This study			Literature studies	
		Avg.	Min-Max	Std.	Sahinkaya et al. (2018)	Dhodapkar et al. (2007)
Cond.	ms/cm	10.45	10.19–10.9	0.90	$9.1 \pm 0.14$	–
COD	mg/L	205.40	85–303	63.40	$208 \pm 18$	70–210
Color	Pt/Co	62.12	46.7–77.70	10.02	$150 \pm 4$	–
TDS	mg/L	6,955	5,154–7,722	0.899	–	7,500–8,500
TOC	mg/L	37.31	15.32–68	16.46	–	–
pH	–	8.48	8.13–9.20	0.05	$8 \pm 0.10$	6–8
Alk.	mg/L*	2,340	2,305–2,380	30.77	$993 \pm 50$	900–1,200
Na <sup>+</sup>	mg/L	2,320	2,162–2,520	116.7	$1,746 \pm 187$	900–1,700
K <sup>+</sup>	mg/L	120	120–128	2.83	$85 \pm 3$	40–100
Ca <sup>2+</sup>	mg/L	71.10	51.9–109.3	20.31	$219 \pm 17$	1,000–1,500
Mg <sup>2+</sup>	mg/L	14.98	9.14–26.30	6.56	$26.5 \pm 7$	400–700
Cl <sup>–</sup>	mg/L	1,510	1,458–1,662	108.64	$1,284 \pm 3$	1,200–2,000
SO <sub>4</sub> <sup>2–</sup>	mg/L	1,920	1,679–2,140	165.53	$2,493 \pm 5$	800–1,200
SiO <sub>2</sub>	mg/L	75.93	47.93–105.87	9.80	$4.64 \pm 0.30$	<0.1
CO <sub>3</sub> <sup>2–</sup>	mg/L	35	30.91–43.90	4.83	–	–
HCO <sub>3</sub> <sup>–</sup>	mg/L	2,310	2,275–2,340	26.64	–	–
Total hardness	mg/L	200	184–357	19.24	$658 \pm 73$	–
NaCl (Salinity)	%	6.17	5.19–6.70	0.37	–	–

Alk, Alkalinity; Avg, Average; Cond, Conductivity (EC); Std, Standard deviation; \*, mg/L as CaCO<sub>3</sub>.

NF270 and NF90 membranes were examined in terms of their removal efficiencies for color, TH, EC, and salinity on the permeate side. The membrane specifications selected for filtration tests are shown in Table 2.

### 2.2.1. Membrane filtration experiments

The crossflow filtration through tubular ceramic membranes contains a peristaltic pump (Filltech, BT 100 2 J), a hot plate and magnetic stirrer (Daihan, MSH-30DSET), a holder, concentrate backpressure valve (1–10 bar), a precision scale (AND, FX-5000i) and a computer, as shown in Figure 2.

The membranes were chemically cleaned before each run of the experiment. The chemical cleaning procedure has two steps: (i) base cleaning for organics removal, and (ii) acidic cleaning for inorganics. Base cleaning was applied with 0.1 N

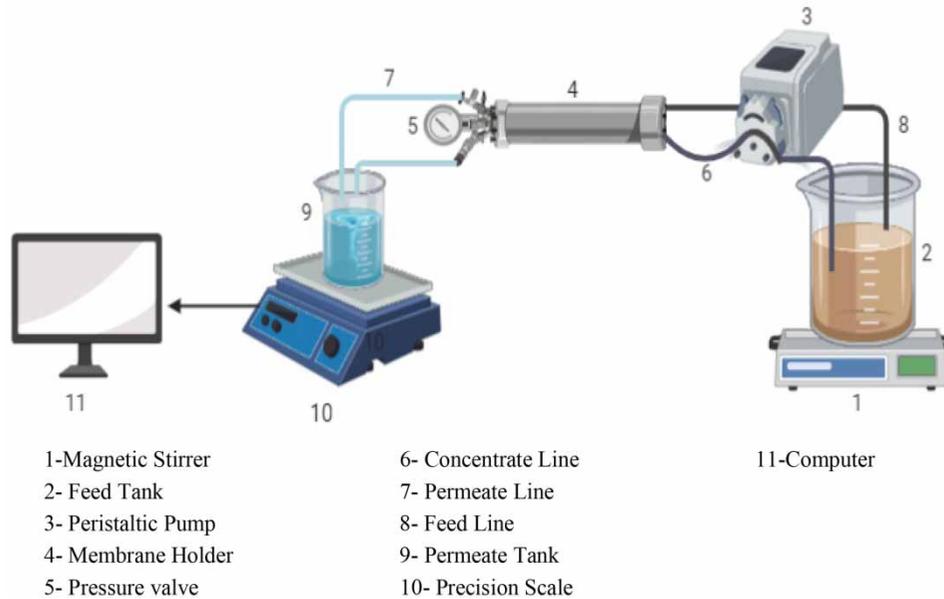
**Table 2** | Examined membranes specification

Parameters	Ceramic Specification	Polymeric	
		NF90	NF270
Support Layer	TiO <sub>2</sub>	–	–
Active layer/polymer	TiO <sub>2</sub> + ZrO <sub>2</sub> (for MF), ZrO <sub>2</sub> (for UF)	Polyamide	Polyamide
MWCO	0,2 μm, 300 kDa, 15 kDa	~200–400 (Da)	~200–400(Da)
Diameter (mm)	10 (outer)–6 (inner)	–	–
Length (mm)	250	–	–
MFA (cm <sup>2</sup> )	40	–	–
pH resistance	0–14	2–11	2–11
OP (bar)	0–10*	25	15
OT (°C) Max	350	45	45
Contact angle	37.92–48.2**	53–57****	27–30****
Rejection (%)	–	99.2 MgSO <sub>4</sub>	99.0 MgSO <sub>4</sub>

\*Membrane holder limitation; MFA, Maximum filtration area; OP, Operating pressure; OT, Operating temperature.

\*\*Ağtaş *et al.* (2021).

\*\*\*Gryta *et al.* (2012).

**Figure 2** | Experimental setup of a tubular crossflow filtration system.

NaOH solutions at 85 °C for 30 minutes and was rinsed until neutral pH with pure water, while acid cleaning was applied using 75% H<sub>3</sub>PO<sub>4</sub> solution (5 ml H<sub>3</sub>PO<sub>4</sub>/L for MF/UF membranes) at 50 °C for 15 min. and was rinsed until neutral pH (Dilaver *et al.* 2018). The operating pressure was maintained at 1 bar for 0.2 μm and 300 kDa and 2 bar for 15 kDa with a concentrate backpressure valve. Crossflow velocity was maintained at  $0.2 \pm 0.03$  m/s using a peristaltic pump during experiments. CMF experiments continued until reaching a recovery rate of 90%. The temperature was kept constant at  $25 \pm 1$  °C using a heating-controlled magnetic stirrer in the experiments. Permeate was collected on a digital balance. Flux (J), and

the normalized flux ( $J_N$ ) of the membrane were calculated using Equations (1) and (2), respectively:

$$J = \frac{V}{A \cdot \Delta t} \quad (1)$$

where  $V$  is the permeate volume (L) for sampling time  $\Delta t$  (h),  $A$  is membrane surface area ( $m^2$ ),  $J$  ( $L/m^2 \cdot h$ ) is the flux at time  $t$  (h) after fouling and  $\Delta t$  is the time difference:

$$J_N = J/J_0 \quad (2)$$

When flux was decreased to about 90% of the initial value ( $J_0$ ), the membrane was taken out and wiped gently with a soft sponge to remove the foulants on the membrane's inner surface. Then the cleaned membrane was used again until the flux could not be recovered by physical cleaning. Permeate was collected on a precision scale and collected permeate was recorded every minute using a computer to calculate the permeate flux (Equation (3)):

$$J = \frac{\left(\frac{\Delta m}{\rho}\right)}{A \cdot \left(\frac{\Delta t}{60}\right)} \quad (3)$$

where  $J$  is the permeate flux [ $L/m^2 \cdot h$ ],  $\Delta m$  represents the collected amount of permeate at specified time interval [g],  $\rho$  is the feed density [ $g/cm^3$ ],  $A$  denotes membrane filtration area [ $m^2$ ], and  $\Delta t$  is time interval [60 s].

Membrane rejection rates are calculated by Equation (4):

$$R(\%) = \left(1 - \left(\frac{C_p}{C_f}\right)\right) * 100 \quad (4)$$

where  $R$  represents membrane rejection for a particular parameter (%),  $C_f$  and  $C_p$  are particular parameters concentration of feed and permeate (mg/L), respectively (Mulder & Mulder 1996).

The Sterlitech HP 4750 stainless-steel stirred cell dead-end filtration unit was used for the NF experiments. The maximum operating pressure of the cell was 69 bar (1,000 psi). Stirring was applied to eliminate concentration polarization with a PTFE stir bar. The cell volume of the system was 300 mL and the membrane filtration area was  $14.6 \text{ cm}^2$ . In the experiment, pure water filtration tests were run before each experiment for compaction. Pre-treated and raw ROC filtration experiments were conducted with an 80% recovery rate. Membrane permeates' flux and rejections rates were calculated using Equations (3) and (4) respectively.

### 2.2.2. Membrane fouling mechanisms (resistance-in-series model analysis)

The membrane fouling mechanisms were studied through the use of the resistance in-series model which illustrates the main fouling mechanism.  $R_m$  which is the hydraulic self-resistance of the clean membrane was identified by measuring the steady-state flux of pure water.  $R_t$  was determined by measuring the steady-state flux of the ROC filtration,  $R_c$  was specified by measuring the pure water flux after physically cleaning the membrane surface and  $R_p$  was specified by pure water flux after chemical cleaning. The sum of the  $R_m$ ,  $R_p$ , and  $R_c$  indicated total resistance to the fouled membrane (Equation (6)).

The resistance was evaluated by using Darcy's Equation (Equation (5)):

$$J = \frac{\Delta P}{\mu \cdot R_t} \quad (5)$$

where  $J$  is the permeate flux,  $\Delta P$  is the applied pressure [Pa],  $\mu$  is the viscosity of the feed [ $Pa \cdot s$ ], and  $R_t$  is the permeation resistance [ $m^{-1}$ ].

$R_t$  which is the total resistance of the filtration system is identified in (Equation (6)):

$$R_t = R_m + R_c + R_p \quad (6)$$

### 2.2.3. Analytical measurements

COD, pH, EC, color,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{SiO}_2$  analysis were performed for the ROC and membrane filtration permeate and concentrate samples. The EC and pH measurements were conducted using the WTW series Inolab pH-EC meter. The COD analyses were carried out using measurement test kits (Hach Lange, Germany) and the Hach Lange DR6000 UV/VIS spectrophotometer. A Thermo reactor system (Spectroquant TR320, Merck) was used for COD analysis. Color (as Pt-Co) analysis measurement was performed according to the Standard Methods (APHA 2005). The TOC was analyzed using a Shimadzu analyzer (Shimadzu, TOC-V CPH model) according to the SM-5310B method. The  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{SiO}_2$  analyses were conducted using an ICP-OES instrument (Perkin Elmer OPTIMA 8300 DV Model) with the ISO-11885 method.

## 3. RESULTS AND DISCUSSION

### 3.1. Coagulation studies

To prevent organic and inorganic fouling on the membrane surface, the ROC was pre-treated using the coagulation process at laboratory scale. The coagulant was selected based on a literature study for similar textile ROC treatment (Umar *et al.* 2016),  $\text{FeCl}_3$  was found to be the best coagulant and optimized with the dosage of  $\text{FeCl}_3$  (1-2-3 and 4 mM) at pH values of 4.0, 5.0, 6.0, and 7.0. The temperature was kept constant at  $25 \pm 1$  °C and  $\text{FeCl}_3$  was added into a jar with 2 L textile ROC while rapidly mixing at 200 rpm for 5 min. It was then mixed at 40 rpm for 15 min. The solution was then settled for an hour and filtered through the 0.2  $\mu\text{m}$  tubular ceramic membrane filtration system. To simulate a CP-CMF process in a laboratory, as is depicted in Figure 2, the optimum dosage was found to be 3 mM at pH 7.

Depending on the type of wastewater; Semblante *et al.* (2018) stated that a range of pH from 3–7 could be used for removal of TOCs, Liu *et al.* (2019) found the optimum pH level to be 5 as well as Ho *et al.* (2015) who declared that pH 7 was optimum in the coagulation process. At neutral pH, removal of TOC,  $\text{Ca}^{2+}$ ,  $\text{SiO}_2$ , and  $\text{Mg}^{2+}$  were noted as 87, 45, 30, and 25%, respectively. At pH values of 7 or higher, there was a small decrease in COD removal. The distribution of iron ionic species may explain this result (Duan & Gregory 2003).

### 3.2. Softening studies

High pH is required for a successful softening process and pH ranges were selected based on literature values. Ordóñez *et al.* (2012) used  $\text{CaCO}_3$  and  $\text{Mg}(\text{OH})_2$  in the range pH of 9.5–10.5 and (Sims 2015) used  $\text{Ca}(\text{OH})_2$  at pH >10. NaOH and  $\text{Ca}(\text{OH})_2$  solutions were investigated to compare their efficiencies and to optimize the required pH adjustment for softening processes in this study. The pH of the samples was adjusted by adding alkaline agents (NaOH and  $\text{Ca}(\text{OH})_2$ ) after 15 min. Of mixing at 200 rpm, and after the desired pH levels were reached, the solution was settled for 1 h and then filtered through CMF.

Use of extreme conditions for precipitation ( $\text{Ca}(\text{OH})_2$  and NaOH), high pH and conductivity values in the effluent were obtained.  $\text{Ca}(\text{OH})_2$  and NaOH processes produced alkalinity in the form of bicarbonates and these processes reduced COD (up to 43.7%) (Prazeres *et al.* 2020). In our case using  $\text{Ca}(\text{OH})_2$  instead of NaOH, enhanced COD removal from 10% to 30%, as is similar to in a study conducted by Latour *et al.* (2014b).

NaOH caused an important increase in the conductivity and salinity of the feed water in contrast with  $\text{Ca}(\text{OH})_2$ , the conductivity of treated water increased by about 0.5 mS/cm similar to the literature result (Semerjian & Ayoub 2003; Latour *et al.* 2014a). Also with the use of NaOH, there was been no significant  $\text{Mg}^{2+}$  and  $\text{SiO}_2$  removal. The optimum pH level was found to be using  $\text{Ca}(\text{OH})_2$  at pH 11.5, in terms of removal efficiency of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{SiO}_2$  (Latour *et al.* 2015).

### 3.3. Silica removal studies

For  $\text{SiO}_2$  removal during the softening treatment, two main mechanisms were regarded: precipitation as calcium and/or magnesium silicate or by adsorption (Latour *et al.* 2015). Precipitation as magnesium silicate is dominant at high pH (Rioyo *et al.* 2018). It was observed that  $\text{SiO}_2$  can be reduced by precipitation as magnesium silicate when the molar ratio of  $\text{Si}/\text{Mg}^{2+}$  is higher than 1/6 (Parks & Edwards 2007). According to the data given in Table 3, the mass ratio of  $\text{Si}/\text{Mg}^{2+}$  was about 2/1 in the ROC, which means sufficient  $\text{Mg}^{2+}$  was not present. Due to the relatively low  $\text{Mg}^{2+}$  content of the ROC,  $\text{Mg}^{2+}$  compounds should be added to increase  $\text{SiO}_2$  removal at high pH. Recent literature studies showed that 80–90%  $\text{SiO}_2$  removal can be obtained by adding  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  and  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$  at a high pH level with high dosages of chemicals (1,500 mg/L) at ambient temperature (Latour *et al.* 2014a). However, adding these  $\text{Mg}^{2+}$  compounds causes an increase in the conductivity of the ROC (Latour *et al.* 2015). In this study, a sparingly soluble  $\text{Mg}^{2+}$  compound ( $\text{MgO}$ ) was used for  $\text{SiO}_2$  removal.  $\text{MgO}$  was tested at four different dosages (from 100 to 400 mg/L) and different pH levels (9.5, 10.5, and

**Table 3** | Concentration results of raw ROC and all treated effluents

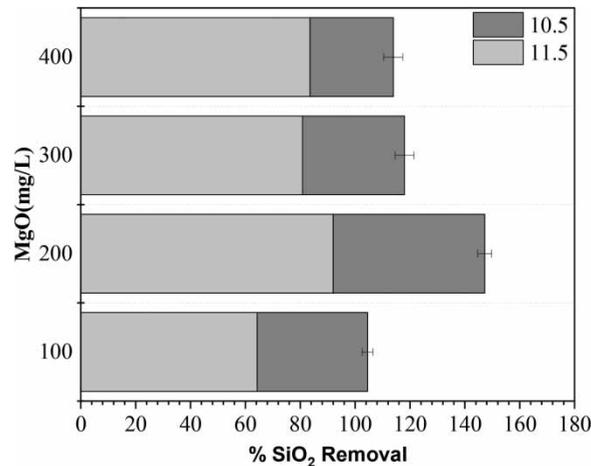
Parameter	Unit	Raw ROC	FeCl <sub>3</sub>	CP-CMF	Water recovery experiments			
					NF90 permeate	NF90 concentrate	NF270 permeate	NF270 concentrate
COD	mg/L	205.40±63.40	61.5±6.75	14.87±5.66	2.75±1.06	389.00±5.30	10.65±0.46	371.00±12.41
Color	Pt/Co	62.12±10.02	17.09±2.23	31.30±3.75	3.40±0.85	133.00±4.47	5.45±0.39	205.00±7.07
Ca <sup>2+</sup>	mg/L	71.10±20.31	45.89±7.25	0.81±0.25	3.19±0.09	4.05±0.40	0.40±0.29	4.11±0.35
Mg <sup>2+</sup>	mg/L	14.98±6.56	12.08±1.12	3.38±0.59	1.675±0.11	19.87±1.02	2.77±0.33	17.22±1.39
Silica	mg/L	75.93±9.80	53.93±8.52	12.99±1.71	<0.50±0.00	66.93±11.73	<0.50±0.00	60.68±9.84
Conductivity	ms/cm	10.45±0.90	10.38±0.05	8.59±0.02	0.73±0.02	37.70±0.18	4.11±0.01	28.67±2.07
Salinity	%	6.17±0.37	6.02±0.04	5.05±0.07	0.37±0.02	23.57±0.10	2.285±0.04	18.85±0.60

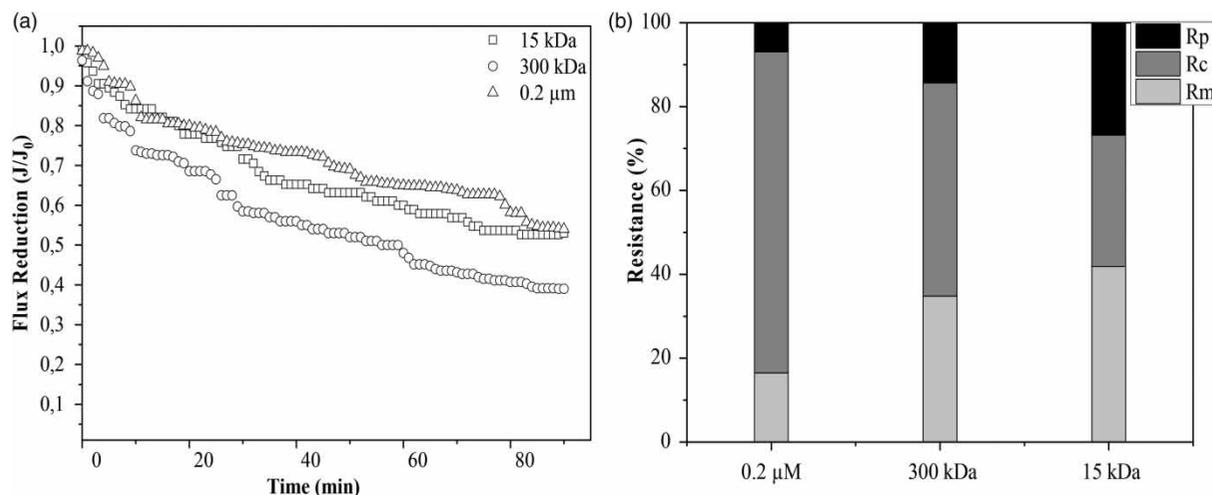
11.5). pH levels and MgO dosages were selected according to previous studies carried out by the most recent literature studies with similar effluent (Sims 2015). Using sparingly soluble compounds of Mg<sup>2+</sup> has the advantage of adding less conductivity to water while causing no further scaling problems. The maximum SiO<sub>2</sub> removal rate was obtained 92% with the addition of 200 mg/L MgO at pH 10.5 and 11.5 as shown in Figure 3, while there was no significant removal efficiency at pH 9.5 from the raw ROC.

### 3.4. CMF studies

The CP-CMF process was conducted in the laboratory as depicted in Figure 2 and 0.2 μm, 300 kDa, and 15 kDa tubular type ceramic membranes were used. All the pre-treatment methods were applied both alone and in combination. The flux reduction fractions ( $J/J_0$ ), was calculated based on the feed flux ( $J$ ) values at a certain time divided by the initial feed flux ( $J_0$ ) value generated during crossflow experiments. The initial and steady-state flux values obtained from the crossflow experiments are presented in Figure 4(a). The flux values that were measured for pre-treated ROC tested with 0.2 μm, 300 and 15 kDa membranes were, respectively, 345±45, 100±10, and 49±2 L/m<sup>2</sup>·h.

When evaluating 0.2 μm filtration studies, which have the highest flux for removal of precipitated divalent ions and organic matter, it was seen that the fluxes sharply decreased within 10 min. and then slowly lessened within the first 80 min. of filtration of the pre-treated ROC (Figure 6(b)). The major reason for this flux decline can be explained by the membrane pore size. In 300 kDa filtration studies 40% flux reduction was observed within 30 min. and total reduction was measured as 65%. For 15 kDa filtration studies, fast flux reduction was observed within 10 min. similar with the other membranes used in this study. This rapid flux decline can be explained by blocking of pores on membrane surfaces by divalent ions and organic matter.

**Figure 3** | Effect of MgO amount for SiO<sub>2</sub> removal (%).



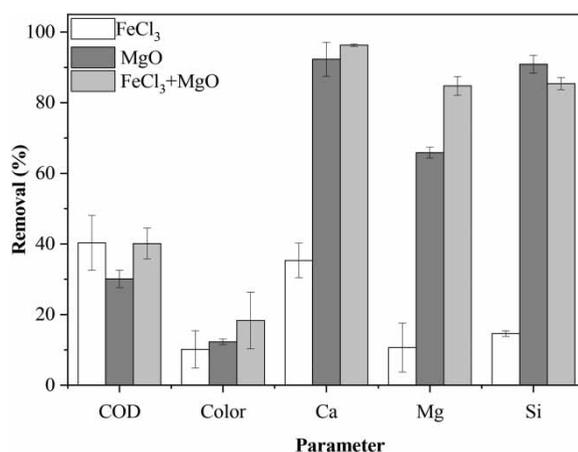
**Figure 4** | (a) Flux behavior during CMF experiments flux reduction, (b) resistances in-series model analysis for CMF experiments.

Membrane fouling mechanisms have been examined with the resistance in-series model during ceramic MF and UF experiments. Generally, the membrane intrinsic resistance ( $R_m$ ) of the membranes increases with decreasing pore size. With this knowledge, the  $R_m$  value of 0.2  $\mu\text{m}$  membranes was found to be lower than 300 and 15 kDa. The  $R_t$  values of 0.2  $\mu\text{m}$ , 300 kDa, and 15 kDa membranes during filtration were observed to be  $7,32 \text{ E} + 11$ ,  $3,93 \text{ E} + 12$ , and  $1,56 \text{ E} + 13$ , respectively, and  $R_p$  values were increased with increasing pore size and found to be  $5,07\text{E}+10$ ,  $5,64\text{E}+11$ , and  $4,19\text{E}+12$ , respectively. The main fouling mechanism was physically removable membrane resistance according to the resistance in-series model for all MWCO of ceramic membranes Figure 4(b). Considering the results, the 0.2  $\mu\text{m}$  value was found to be more appropriate with a high flux rate and lowest membrane resistance values among the three membranes tested for pre-treatment.

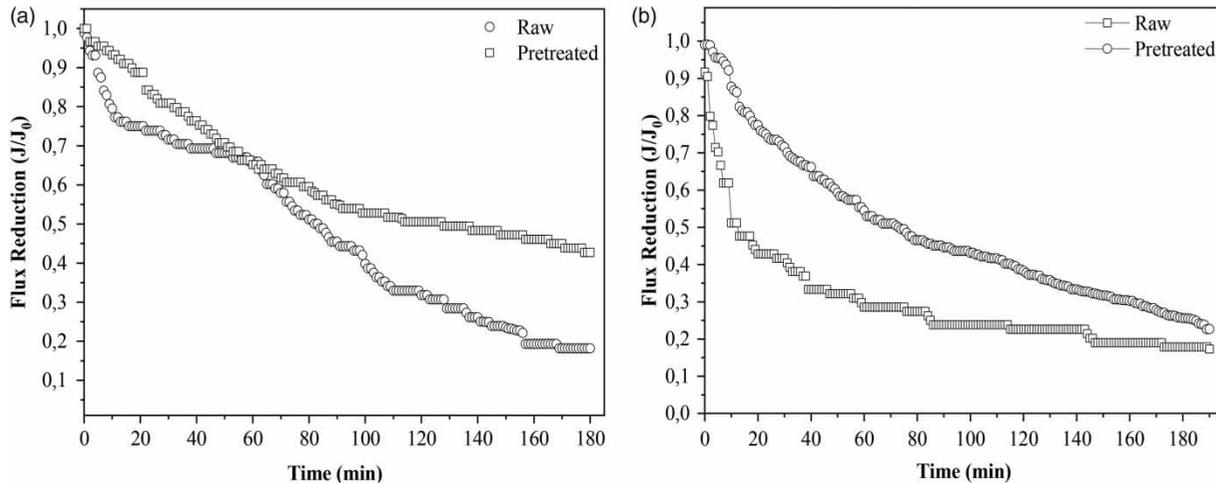
### 3.5. CP-CMF results

At neutral pH using 3 mM  $\text{FeCl}_3$ , the requisite amount of  $\text{Ca}(\text{OH})_2$  for achieving the targeted softening of pH and 200 mg/L MgO addition to the ROC, the 0.2  $\mu\text{m}$  pore size of CMF was found as the best pre-treatment. The  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{SiO}_2$  removal performance by  $\text{FeCl}_3$  was found to be 45, 23, and 30%, respectively, while 88, 71, and 92% removal was observed, respectively, with 200 mg/L MgO at pH 11.5. Adding MgO compound to ROC increased  $\text{SiO}_2$  removal almost threefold (Figure 5). All pre-treatment methods were conducted using the CMF and were repeated at least duplicated.

A study was conducted with a UF membrane integrated pellet reactor that can remove scale-forming ions such as  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and Si from a pilot-scale textile industry ROC. The resulting effluent was further tested in a secondary RO process



**Figure 5** | Comparison of removal efficiencies of conducted CP-CMF experiments.



**Figure 6** | Pre-treated ROC flux reduction during (a) NF90 and (b) NF270.

to reduce the amount of concentrate and increase overall water recovery. Unlike raw concentrate, after pellet treatment, more than 80% water recovery was provided without a dramatic flux decrease (Sahinkaya *et al.* 2018). In another study, resulting from the desalination unit, ROC was introduced to a ceramic UF membrane following chemical pre-treatment for enhancement total water recovery. A CUF membrane were used for increasing solid concentration and minimizing membrane concentrate (Sanciolo *et al.* 2012).

Technically, using ceramic MF and UF as pre-treatment steps before NF and RO is the optimum option for textile wastewater with high COD/BOD and TDS concentrations (Samaei *et al.* 2018).

### 3.6. ROC minimization and water recovery studies

NF membranes such as NF90 and NF270 have a tight polyamide separating layer (Zhou *et al.* 2015) and high rejections of polyvalent ions and low-to-moderate rejections of monovalent ions and small organic compounds (Lin *et al.* 2017). For salt removal compared to RO, NF90 might be the best option (Hilal *et al.* 2005; Zhou *et al.* 2015) as it ensures the key differentiating characters of NF are their higher flux, lower energy consumption, and longer membrane life (Mohammad *et al.* 2015). For these reasons, NF filtration tests were conducted with NF90 and NF270 membranes, and pre-treated samples were filtered through both membranes at least duplicated. A new membrane was used for each of the filtration stages and rinsed with pure water before the experiment and pre-treated ROC samples pH adjusted to 7–8 before the NF filtration experiments.

Raw ROC and pre-treated ROC concentrations after each treatment method are given in Table 3. Evaluating NF270 permeates, medium silica, Ca<sup>2+</sup>, and Mg<sup>2+</sup> removal were observed. The NF concentrate showed a lack of Ca<sup>2+</sup>, the same average concentration of Mg<sup>2+</sup> was found in the raw ROC, and silica values six times lower than the raw ROC concentration. While the NF90 permeate shows almost no organic pollutant and silica content, the low conductivity (0.74 ms/cm) and low salinity (0.38%) concentrations allow the permeate to be reused in the process. The NF90 concentrate COD, color, Ca<sup>2+</sup>, Mg<sup>2+</sup>, and silica concentration were measured as 389, 133, 4.05, 19.87, 31.12 mg/L, respectively, and, theoretically, recirculating NF concentrate is possible.

For MgO precipitation combined with NF270 filtration, the removal efficiencies were found to be: COD, color, Ca<sup>2+</sup>, Mg<sup>2+</sup> and SiO<sub>2</sub>; 92, 83, 97, 84 and 92%, respectively. Using MgO + NF90 tests, the removal efficiencies were COD, color, Ca<sup>2+</sup>, Mg<sup>2+</sup> and SiO<sub>2</sub>; 95, 85, 97, 81 and 98, respectively and lastly, for CP-CMF/NF90 tests, the removal efficiencies were COD, color, Ca<sup>2+</sup>, Mg<sup>2+</sup> and silica, 98, 94, 97, 93, 99%, respectively (Figure 6(a)). When CP-CMF + NF270 tests were carried out, the removal efficiencies were COD, color, Ca<sup>2+</sup>, Mg<sup>2+</sup> and SiO<sub>2</sub>, 97, 89, 96, 87 and 98%, respectively (Figure 6(b)). The most efficient pre-treatment method was determined to be integrated FeCl<sub>3</sub> + MgO adding CMF. For further treatment, NF90 showed higher removal efficiency for COD, color, total hardness, and silica than NF270. Alongside decreased salinity and conductivity and increased permeate rates. NF90 membranes were found to be the best option for water recovery. These membranes were also examined for flux behavior in addition to their removal efficiencies, Figure 6 shows the steady-state flux values obtained after filtration tests for raw and pre-treated ROC for both two NF membranes.

The highest steady-state flux value for all membrane tests was obtained with the NF270, which has a large pore size. The lowest steady-state flux values for the two membranes were obtained after filtration tests with raw ROC. A significant increase in steady-state flux value for the NF270 membrane was obtained from the filtration tests after CP-CMF was used, while the steady-state flux of NF90 membrane increased approximately 1.5 times after the pre-treatment processes. While the fastest flux decline rate for the NF270 filtration test was measured with the pre-treated sample as 50% within the first 50 min., this rate was 70% with raw ROC. When the NF90 filtration tests were evaluated, within the first 50 min., 40% flux decline was observed for both pre-treated and raw ROC; after 150 min filtration, 50% flux decline was measured for the pre-treated ROC while the raw ROC flux decline was measured to be 75%.

In the NF90 filtration tests, no significant differences were found using  $\text{FeCl}_3$  and MgO in terms of flux but their combination increased the flux 1.5 times. In NF270 filtration tests, with the addition of MgO, the scaling problem was eliminated at pH 11.5, and flux rate increased 1.5 times; for pre-treatment with  $\text{FeCl}_3$  + MgO, this ratio was doubled (Figure 7).

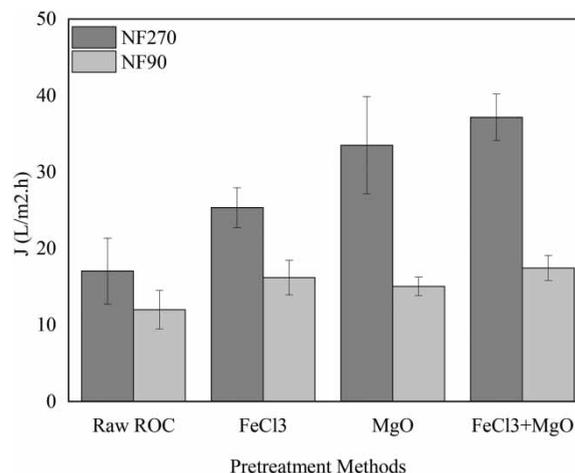
### 3.7. Water reuse perspectives

A certain objective of the study was the utilization of ceramic membrane technology in order to achieve the standards recommended for reuse in textile processes. Addressing the quality of purified water for reuse purposes, it has to be noted that the specific requirements can be quite different according to a company's needs. Nevertheless, certain general criteria have been established that should be met by the treated water (Li & Zhao 1999; Rozzi *et al.* 1999). These are: COD <30 mg/l, a spectral absorption coefficient (SAC) at 426 nm <1.0 ( $\text{m}^{-1}$ ) and conductivity <1.8 mS/cm (Table 4).

The suggested pre-treatment process is a reasonable treatment option in combination with other advanced treatment technologies to achieve reuse standards. In particular, the permeate is suitable to be directly fed to the nanofiltration module. This two-stage process configuration has been tested and produces a water quality suitable for any textile process.

### 3.8. Cost estimation

Cost estimation was carried based on optimum conditions determined in the proposed experimental studies. The plant was postulated to operate for 24 h per day and 365 days per year. The costs of the components were determined from market values and adapted from the available literature. For the cost analysis, the investment cost of ceramic membranes is significantly higher; the lifespan is on average of 15–20 years, approximately doubled their polymeric counterparts (Gitis & Rothenberg 2016). Based on the availability of real-scale data, the costs for CMF installation and operational costs were assumed as follows. Ceramic membrane costs are about 1,500–2,000 USD/ $\text{m}^2$  and the module cost is 16–32 USD/ $\text{m}^2$  (Dilaver *et al.* 2018). To treat 480  $\text{m}^3/\text{d}$  of water with 250 LMH, the required membrane area is 80  $\text{m}^2$  which accounts for 120,000 USD for the membranes and approximately 2,000 USD for the membrane module. In a recent study, polymeric NF and RO membrane cost was assumed to be 40 USD/ $\text{m}^2$  (Panagopoulos 2021). Pannirselvam *et al.* (2019) stated that the price of NF membranes was between 30–60 USD/ $\text{m}^2$  with an average of 45/ $\text{m}^2$  within their economic analysis studies for NF membrane costs (Pannirselvam *et al.* 2019). Vergili *et al.* (2012) estimated the NF membrane cost for integrated membrane



**Figure 7** | Flux values for each pre-treatment method for NF90 and NF270.

**Table 4** | Textile wastewater reuse criteria

Parameters	BTTG, 1999	González-Zafrilla et al. (2008)	Yin et al. (2019)	Rozzi et al. 1999 Reuse in reactive and dyeing processes synthetic fiber dyeing	Li & Zhao 1999 Reuse in reactive and dyeing processes
COD (mg/L)	80	< 20	60	30	0–160
TSS (mg/L)	5	–	30	–	–
TDS (mg/L)	500	–	–	–	–
Hardness (mg/L as CaCO <sub>3</sub> )	60	–	450	–	–
Conductivity (μS/cm)	1,000	500	–	1,800	800–2,200
Color	20(Pt–Co)	–	30(Pt–Co)	0.01(426 nm)	0–2 Lovibond
pH	6–8	–	6.5–8.5	–	–
Turbidity (NTU) 1	1	–	5	–	–
Cl <sup>-</sup> (mg/L)	–	–	250	–	–
NH <sub>3</sub> -N (mg/L)	–	–	30	–	–

scenarios as 30 USD/m<sup>2</sup> (Vergili et al. 2012). In our country market prices for NF membranes are about 50–100 USD/m<sup>2</sup>. Thus far, there is no information on the cost of ROC treatment by coagulation. In this case, the costs were found for chemicals to be 0.26 USD/m<sup>3</sup> (the total cost of used FeCl<sub>3</sub>, MgO, and Ca(OH)<sub>2</sub> per m<sup>3</sup>). Disposal of chemical pre-treatment sludge cost was included in the total cost of CMF concentrate disposal adapted from Heijman & Bakker (2007). A summary of expenditures is given in Table 5. For CMF, the operational, specific, and maintenance costs were adapted from the TECHNEAU Report Heijman & Bakker (2007), a recent study conducted by Weschenfelder et al. (2016), Viegas et al. (2020), and NF costs adapted from Zhou et al. (2015). The costs of mixing and pumping energy requirements corresponding to 0.026 kWh/m<sup>3</sup> for the CP-CMF process were included (Tompkins et al. 2019), in a recent work with ceramic MF membrane energy consumption was given as 0.4 kWh/m<sup>3</sup> (Hakami et al. 2020), in our study it was calculated as 0.15 kWh. The energy consumption of

**Table 5** | Cost-benefit analysis and operational and capital cost of the proposed study

CMF	NF90		
	USD/m <sup>3</sup>	USD/m <sup>3</sup>	
Depreciation, energy, chemicals, maintenance and concentrate disposal are approximately	0.14	Initial investment membrane and holder	0.10
Chemical pre-treatment cost	0.26	Energy cost (2.2 kWh/m <sup>3</sup> )	0.15
Initial investment membrane and holder	0.03	Membrane replacement	0.05
Membrane replacement	0.07	Regeneration cost	0.03
Labor	0.14	Total NF cost	0.32
Energy consumption (0.15 kWh/m <sup>3</sup> )	0.01	Proposed total cost	0.97
Total cost for CMF	0.65	Total water saving	0.50
		Total cost	0.47
Wastewater discharge price	0.20 USD/m <sup>3</sup>	Fresh water price	0.30 USD/m <sup>3</sup>
Energy cost for pumping and mixing	0.07 USD/kWh	Personnel costs (annual cost)	8,000 USD/worker-y
FeCl <sub>3</sub>	0.50 USD/kg Fe	MgO	0.11 USD/kg
Ca(OH) <sub>2</sub>	0.20 USD/kg		

\*Provided by the suppliers from the market; average value of Turkey.  
Maintenance 1.5% of the total capital costs

the NF process is about 0.245 kWh/m<sup>3</sup> by (Turek *et al.* 2009), in another study it was assumed to be 1.4 kWh/m<sup>3</sup> for NF90 (Schäfer *et al.* 2007). In this study it is assumed to be 2.2 kWh. Major operating costs, the electricity, labor, and chemical consumptions costs are presented in the Supplementary information.

Annually, 175,200 m<sup>3</sup> water will be saved through the proposed study. Both required wastewater treatment and fresh/process water consumptions will be reduced, and 87,600 USD are saved due to water recovery. The total net cost of the proposed study with the consideration of savings is about 0.47 USD/m<sup>3</sup> (Table 5).

ROC management processes were evaluated; it was applied for treating textile RO concentrate using ED and evaporation alone and a combination of these processes. Only an evaporation process was used for ROC treatment and the cost of the method was found to be 3.88 USD/m<sup>3</sup>, while the ED and evaporation combination is approximately 0.55 USD (Praneeth *et al.* 2014). Using the ASP process it was 2.6 USD/m<sup>3</sup> (Sanciolo *et al.* 2012), using RO–NF combination process cost was 0.57 USD/m<sup>3</sup> (Chen & Chen 2004). A combination RO + ED + Crystallization method was conducted by Nayar *et al.* (2019) and the cost of the proposed method was 3.5 USD/m<sup>3</sup> (Nayar *et al.* 2019). Şahinkaya *et al.* calculated the treatment cost of the pellet reactor as 0.83 USD for 1 m<sup>3</sup> ROC (Şahinkaya *et al.* 2018). UV, ozonation, and evaporation processes were discovered to be the most expensive methods. Compared with the other methods, CP–CMF/NF may be one of the cost-effective processes for water recovery.

#### 4. CONCLUSION

The pre-treatment method of this study is suitable in terms of removal efficiency and enhancement of flux rate. It was able to precipitate scaling cations and remove organic matters from textile industry ROC before the NF process. For further treatment, NF90 and NF270 membranes were used to reach desired water recovery values. The results show that NF90 was the effective NF membrane compared with NF270 in achieving desired removal efficiency and flux rate of almost 50%. Moreover, the NF concentrate analysis indicated that recirculation of the concentrate stream is technically possible. From an economic viewpoint, with the proposed method the return of the initial investment is expected within 2 years. As compared to the other methods, CP–CMF/NF may be a cost-effective approach and freshwater consumption can be reduced by 15% and the requirements for water reuse may be met. The CMF integrated CP process has the potential for pilot-/real-scale applications employing RO processes, municipal/industrial wastewater treatment, and reuse to achieve near-ZLD by CP–CMF/NF.

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#### DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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