



Hybrid coagulation–NF membrane processes for brackish water treatment: Effect of pH and salt/calcium concentration



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HIGHLIGHTS

- Hybrid coagulation–NF membrane process for brackish water desalination was studied.
- Influence of water chemistry on the performance of hybrid process was investigated.
- Properties of the foulants in the solution were altered by coagulation process.
- Distinct membrane fouling phenomena compared to single fouling study were observed.
- Membrane foulant cake layer was inspected and linked to explain fouling mechanisms.

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ABSTRACT

In order to reduce membrane fouling propensity, membrane technologies in water treatment and desalination processes have been coupled with coagulation process. However, the fouling issues still persisted. It is believed that coagulation might affect the fouling mechanisms involved. Hence, this study was intended to investigate the influence of water chemistry (solution pH, calcium concentration, and total dissolved solids) on hybrid coagulation–nanofiltration membrane processes. Assessment will be done based on permeate flux, salt rejection trend, and foulant layer autopsy. It was discovered that coagulation process altered the properties of the foulants in the solution and resulted in distinct membrane fouling phenomena compared to most of the standalone membrane fouling studies. This situation prevailed in the presence of calcium ions in the solution. It was postulated that coagulation process weakened the interaction between the oppositely charged calcium ions and foulants. Therefore, instead of forming complex with the foulants, calcium ions had higher affinity towards more negatively charged membrane. Consequently, calcium salts precipitated and formed scales on the membrane surface, as supported by membrane performance and SEM-EDX analysis. This indicated that membrane fouling study should consider the impact of pretreatment process for better understanding about the fouling mechanisms involved.

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1. Introduction

Today, membrane filtration technology is gaining momentum in water treatment processes and it is driven by two major factors: poor raw water resources and stringent drinking water policy. In addition, membrane technology offers several advantages over the conventional water treatment processes, such as lesser chemical consumption, better water quality, and smaller plant footprint [1]. However, fouling which

degrades the membrane performance is one of the major challenges in membrane applications for water industry. Hence, various studies related to membrane fouling have been carried out actively, with most of the interest focus on the influence of water chemistry on the membrane performance especially in rejecting natural organic matter (NOM). Water chemistry such as pH and the composition of the dissolved mineral ions in the water has significant impact on the membrane performance [2,3]. For instance, low pH and high ionic strength aggravated membrane fouling by reducing the electrostatic repulsion between the foulants and membrane. The formation of compact foulant layer will block the passage of water through the membrane [4,5]. Besides surface charge, properties of membrane surface may be altered by water chemistry. Kilduff et al. [4] reported that when membrane charge was

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neutralized (acidic and high ionic strength conditions), reduced membrane permeability may be contributed by the reduction in membrane porosity or pore size. However, Nanda et al. [6] observed that membrane flux was enhanced while salt retention capability was reduced at low pH and they attributed this to the thinning of active layer on the nanofiltration membrane surface under acidic medium. Such contradicted findings may arise from the type of membranes used in the study.

To date, most of the reported fouling studies have been done on standalone membrane process and with synthetic foulants such as humic acid (HA) [2–5,7,8]. In real industry application, normally there will be some pretreatment processes prior to the membrane filtration unit, such as coagulation, adsorption, preoxidation, and biological treatment [9,10]. Pretreatment is designed to remove part of the foulants in raw water and to reduce the fouling propensity of the membrane unit. However, pretreatment process which involves the addition of chemicals such as coagulation may alter the properties of the foulants in the solution [11,12]. These changes may affect the membrane performance and fouling propensity. As the potential and benefits of hybrid coagulation–membrane process has been reported and its implementation is gaining ground, membrane fouling study in this field should as well receive more attention [13]. Hence, a systematic fouling study should be carried out to investigate and compare the influence of water chemistry on the performance of hybrid coagulation–membrane process. Such investigation can provide a better understanding about the fouling mechanisms involved in the hybrid coagulation–membrane process.

Membrane fouling has been widely reported in relation to the formation of foulant cake layer on the membrane surface [6,14–18] and represented by the decline of membrane permeability and salt rejection capability [4,19–21]. To our best knowledge, formation of foulant cake layer on the nanofiltration membrane surface under the influence of water chemistry has not been well investigated. Similarly, very few works have tried to relate the fouling trend with cross sectional view of the fouled membrane to support their fouling mechanism postulation. Hence, it will be interesting to link and confirm the fouling postulation with the FESEM results of the cake layer.

This study aims to investigate the influence of water chemistry on the fouling propensity of NF membrane process receiving supernatant solution from the coagulation pretreatment process. Synthetic solutions with different salt compositions close to the real water compositions will be used. Nanofiltration membrane (NF 270) which is suitable for brackish water treatment will be used in this study. By using such approach, additional fouling information can be used in combination with other postulations to have better understanding about the hybrid coagulation–membrane fouling issue under the influence of water chemistry.

2. Methodology

2.1. Chemicals and membranes

All chemicals used are analytical grade, unless stated otherwise. Humic acid (HA), ferric chloride (FeCl_3), kaolin, calcium chloride ($\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$), sodium bicarbonate (NaHCO_3), and sodium chloride (NaCl) were purchased from Sigma Aldrich (Malaysia). Ultrapure (UP) water with a quality of $18 \text{ M}\Omega \text{ cm}^{-1}$ was used for all solution preparation. Membrane used in this study was NF 270 purchased from Dow

Filmtec (USA). The characteristics of the membranes are shown in Table 1.

2.2. Synthetic test waters

Synthetically prepared waters with fixed turbidity were used for this work. The HA concentration for each batch of run was 20 ppm. Suitable amount of kaolin was added into the synthetic water to adjust its turbidity to $30 \pm 0.5 \text{ NTU}$. The pH of all the synthetic water prior to coagulation process was adjusted to 7 by using sodium hydroxide (NaOH) and hydrochloric acid (HCl). The zeta potentials of the synthetic waters with different total dissolved solids (TDS) were shown in Table 2. W5 and W6 solution were the supernatant solutions from W1 and W3 where the pH was adjusted to 7.

2.3. Jar test coagulation and cross-flow process setup

Coagulation pretreatment prior to NF membrane process was carried out in a conventional jar test apparatus (Model ZR4-6, Zhongrun Water, China). The coagulation procedures consisted of three steps: vigorous stirring after the addition of coagulant (100 rpm for 1 min), mild stirring (30 rpm for 29 min), and settling (30 min). The dosage of FeCl_3 coagulant was varied in order to obtain the optimal dosage which removed most of the turbidity and HA. The supernatant from the coagulation process with optimal dosage was then used as the feed water for membrane experiment.

Bench-scale cross-flow membrane experimental setup with recycle loop as shown in Fig. 1 was used for this experiment. The membrane test cell (CF 042, Sterlitech, USA) has 0.0042 m^2 membrane effective filtration areas. The supernatant water from the coagulation process will be used as the feed for the cross-flow system. The operating conditions for temperature, pressure, and flow rate were 27°C , 10 bars, and 42 cm/s respectively. This membrane filtration experiment was conducted for 5 h with all the operating conditions being controlled and maintained at the values aforementioned. The performance of the membrane process was assessed and presented as flux versus time and salt rejection versus time.

2.4. Analytical methods

Humic acid absorptivity was measured using a UV/Vis spectrophotometer (PerkinElmer, USA) at a wavelength of 254 nm. Zeta-Sizer (Malvern, UK) was used to measure stability of the suspensions. Turbidity of the water was measured using a 2100 N Laboratory Turbidimeter (Hanna, USA). Conductivity and pH of the solution were measured using a HI 2550 Benchtop Meter (Hanna, USA). The fouled NF 270 membranes were characterized (surface view and cross sectional view) using field emission scanning electronic microscopy–energy dispersive X-ray (Merlin Compact, Zeiss, Germany).

3. Results and discussion

3.1. Characteristics of the supernatant solution after coagulation process

Zeta potentials of the raw synthetic waters as presented in Table 2 showed an obvious difference for solution with calcium ions and alkalinity. Foulants in W2/W4 solutions have much lower zeta potential compared to W1/W3 solutions. This reflected that the presence of

Table 1
Characteristics of NF 270 membrane used in the study.

Membranes	Molecular weight cut-off (MWCO) (Da) ^a	Root mean square (RMS) roughness (nm) ^b	Zeta potential (mV) ^b
NF 270 (NF)	200–300	9.0 ± 4.2	–32.6

^a Data provided by manufacturer.

^b Zeta potential values (at pH 7) and surface roughness were taken from [22].

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