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Hybrid coagulation–NF membrane process for brackish water treatment: Effect of antiscalant on water characteristics and membrane fouling

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HIGHLIGHTS

- Scaling could severely impair the performance of membrane desalination process.
- Antiscalants have been used to prohibit and reduce the risk of scale formation.
- Influence of antiscalant on coagulation–NF membrane process was investigated.
- Interaction between antiscalant and foulants varied according to its concentration.
- Membrane fouling tendency and water characteristic changed with antiscalant dosage.

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ABSTRACT

Antiscalants have been used to inhibit the formation of scales and reduce the risk of membrane scaling. However, issues such as the increased of biofouling tendency and contradictory observations of organic fouling propensity under the influence of antiscalants have been reported by other researchers. Such observations indicate the need for thorough understanding about the interaction between antiscalants and membrane/foulants in the solution. This study investigated the influence of antiscalant on the water characteristics and the performance of hybrid coagulation–nanofiltration membrane process for brackish water treatment. It was observed that scaling was mitigated with the addition of antiscalants but membrane fouling was exacerbated at higher dosage, probably due to the changes in foulant properties induced by antiscalants. Analysis carried out on the water showed that antiscalants altered the characteristics of the foulants and formed precipitates with calcium/aluminum and iron (from coagulant) ions, which eventually deposited as foulant layer on the membrane surface. This was proven with the existence of phosphorous element on the membrane surface using SEM–EDX. The findings demonstrated how the antiscalants altered the water characteristics and provided insight into the reactivity of antiscalants present in different concentrations, which could be used to interpret the contradicting findings from other researchers.

1. Introduction

Scaling is a severe problem for membrane desalination process because it could impair plant performance, reduce permeate quality, and cause economic loss due to shorter membrane lifespan, more frequent chemical cleaning, and loss in water production [1]. High dissolved salt retention by nanofiltration (NF) and reverse osmosis (RO) membranes results in concentrated dissolved salts at the membrane surface. Eventually the dissolved salts start to precipitate once its solubility level has been exceeded. Scales such as calcium sulfate, calcium carbonate, and barium sulfate are the typical inorganic salts that precipitate on the membrane surface [2]. In order to alleviate scaling issues, controlling strategies such as pH adjustment, antiscalant addition, and chemical cleaning have been commonly applied in membrane desalination process [3–5].

Dosing of antiscalants prior to the membrane filtration unit to inhibit the formation of scales appears to be the most recommended approach because of its advantages; operational and capital cost reduction, environmental acceptability, and safety compared to other alternative technologies [6,7]. Antiscalants may interfere with the formation of scales during scale nucleation and crystallization

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processes via several approaches such as threshold inhibition, crystal modification, dispersion, and chelation [8–11]. Even though antiscalants play a vital role in minimizing the scaling risk of membrane desalination processes, several studies have reported that the application of antiscalants contributed towards other fouling issues. Increased biofouling propensity has been linked to the use of antiscalants, probably by serving as an additional source of phosphorous (nutrient) to the microbial population in the seawater [12–14]. Furthermore, relations between organic foulants and antiscalants are complicated as some studies found that antiscalants enhanced organic fouling while on the other hand, it was reported that organic fouling might be mitigated under appropriate control of antiscalant dosage [1,8,15,16]. Also, it has been reported that agglomeration may occur between the anionic antiscalants and cationic flocculants used in the pretreatment stages and may cause membrane fouling issue [17,18]. Similarly, effectiveness of antiscalants can be reduced when antiscalants react with polyvalent cations like Fe^{3+} and Al^{3+} [5,11,17]. Hence, it can be seen that the interactions between antiscalants and components in water are complicated and further research on this issue should be explored.

Standalone membrane process is unsustainable due to its high fouling risk. Hence, pretreatment prior to the membrane unit is inevitable because it can significantly reduce the risk of fouling and enhance the performance of the membrane process [19]. Such hybrid/integrated membrane processes have been widely applied in real desalination plants. Coagulation is one of the most popular and efficient pretreatment processes where coagulants are added into the raw water to remove suspended solids and turbidity via several mechanisms such as charge neutralization and sweep coagulation [20,21]. The interaction can be done because coagulants are usually positively charged and foulants are normally negatively charged. Thus, there is a probability that the leftover cationic coagulants will travel to the membrane filtration unit and interact with anionic charge antiscalants. The complication might produce substances that have adverse effect on the membrane performance. However, scientific details about this have been scarce and such study has not been widely reported before.

NF membrane is very effective in rejecting divalent dissolved salts which normally make up a large portion of dissolved salts in brackish water. Furthermore, NF membrane requires lower operating pressure and produces higher flux. This makes it a potential alternative to RO membrane in treating brackish water with lower salinity [22,23]. As far as our concern, there has been no reported study on the effect of antiscalants on hybrid coagulation–NF membrane process. Hence, this study aims to investigate the impact of antiscalants on the performance and fouling propensity of hybrid coagulation–NF membrane process in treating brackish water. The dosage of antiscalants will be varied and the change in water characteristics and membrane performance will be analyzed. A study on the change in water characteristics under the influence of high antiscalants dosage can provide further understanding about the possible interactions between antiscalants and substances in the solution. Understanding about the relationship between the excess antiscalants and foulants in the solution can be utilized and linked to the membrane performance. It is postulated that antiscalants will modify the properties of the foulants in the water as well as the characteristics of the membrane surface, which in the end affect the overall performance and fouling propensity of the membrane separation process.

2. Methodology

2.1. Chemicals and membranes

All chemicals used are of analytical grade, unless stated otherwise. Humic acid (HA), ferric chloride ($FeCl_3$), kaolin, calcium chloride ($CaCl_2 \cdot 2H_2O$), sodium bicarbonate ($NaHCO_3$), and sodium chloride (NaCl) were purchased from Sigma Aldrich (Malaysia). Ultrapure (UP) water with a quality of 18 MΩ 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. Antiscalant used in this study was Tripol 8510 (Trisep, USA). It consists of approximately 10% of polyacrylic acid and diphosphonic acid each [24].

2.2. Synthetic test waters

Synthetically prepared brackish 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 ± 0.5 NTU. The pH of the water was adjusted to 7 by using sodium hydroxide (NaOH) and hydrochloric acid (HCl). The zeta potentials of the synthetic waters with two different total dissolved solids (TDS) were shown in Table 2.

2.3. Jar test coagulation and cross-flow process setup

Coagulation pretreatment prior to NF/RO 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. Before the cross-flow experiment was started, antiscalant was dosed into the feed solution. The dosage of antiscalant (shown in Table 3) can be divided into low dosage (A and B) and high dosage (C–E). Low dosage was used to observe whether membrane scaling can be prevented by the addition of antiscalant. High dosage was applied to investigate the fouling induced by the addition of antiscalant. Dosage of antiscalant as tabulated in Table 3 for the membrane process was selected in this study because the pH of the supernatant solution decreased drastically under much higher concentration of antiscalant, which made it inappropriate for the operation of membrane process. The operating conditions for temperature, pressure, and crossflow velocity are 27 °C, 10 bars, and 42 cm/s respectively. The 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 absorbance was measured using a UV/Vis spectrophotometer (PerkinElmer, USA) at a wavelength of 254 nm. A Zeta-Sizer (Malvern, UK) was used to measure the stability of the suspensions; and a Master-Sizer (Malvern, UK) was used to determine the particle

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 [25].