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Application of chlorine dioxide (ClO_2) to reverse osmosis (RO) membrane for seawater desalination

Jung Hwan Oh, Am Jang*

Graduate School of Water Resources, Sungkyunkwan University (SKKU), 2066, Seobu-ro, Jangan-gu, Suwon-si, Gyeonggi-do, 16419, Republic of Korea

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ABSTRACT

There are justifiable concerns about reverse osmosis (RO) performance deterioration and carcinogenic by-products from disinfection in water treatment facilities. Traditional disinfectant chlorine (Cl_2) has been replaced with stable and safe disinfection processes. Chlorine dioxide (ClO_2) as an alternative disinfectant has been increasingly adopted in various water treatment facilities. To identify the applicability of ClO_2 to RO desalination process, lab-scale experiments were designed by simulating actual oxidant exposure circumstances. In the experiment, Cl_2 or ClO_2 solution was injected separately into a working RO system. Our results showed that Cl_2 dosage greatly decreased permeate flux slope. However, instant flux increase of approximately 5% was observed when ClO_2 was injected. ATR FT-IR and EDX analysis for exposed membranes showed that Cl_2 deformed amide peaks and attached Cl atom onto the membrane, indicating chlorination. However, none of these results was observed in ClO_2 exposure cases. In addition, immediate sodium bisulfite application alleviated flux decrease under Cl_2 chlorination.

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1. Introduction

Disinfection mechanisms and disinfectants are of great concern because they could affect various fields, including the production industry, water purification, and wastewater treatment. Disinfection also plays a significant role in desalination, especially in pre- and post-treatments designed to inactivate microorganisms in source water and preserve treated water [1]. Chlorine (Cl_2), a strong oxidizing agent, has been widely used as a disinfectant in the twentieth century because of its important characteristics such as high toxicity to microorganisms, high deodorizing ability, stable usage, and reasonable cost [2]. Specifically, available chlorine and time multiplied value (C·T) have been widely used as indices to carefully monitor the disinfection process and prevent bio-fouling due to microorganisms to meet water quality standards [3–5].

However, using Cl_2 as a disinfectant has negative side effects. Many studies have warned about the environmental toxicity of disinfect by-products (DBPs) formed by reactions with organic/inorganic matters in the Cl_2 disinfection process [2,5–11]. Moreover, due to public concerns about carcinogenic compounds, certain DBPs including total trihalomethane species and five haloacetic acids have been strictly controlled and regulated in the United States [12].

Other concerns related to Cl_2 disinfection are at the reverse osmosis (RO) desalination process. Although various reducing agents are produced for the dechlorinating process followed by chlorination, partial failure of that process can expose RO membranes to residual Cl_2. It has been proven that even a low concentration of free Cl_2 can deteriorate the polyamide membranes and deform the amide functional groups, resulting in irreversible decrease in water flux during long-term operation [13–16]. Moreover, several studies have casted doubts on the security of RO in DBPs filtration. For instance, although RO can filtrate most solutes, its rejection of chloroform and bromoform is unreliable [17,18].

Since the introduction of chlorine dioxide (ClO_2) as an alternative disinfectant in the late twenties century, it has been adopted in various water treatment facilities. This is due to the featured characteristics of ClO_2 such as having equal to or greater disinfecting power compared to that of Cl_2, more effective in deactivating viruses than Cl_2 [2], and producing no harmful DBPs when reacted with organic matters [19]. In seawater RO process, however, there are few studies concerning ClO_2 as an alternative disinfectant. A previous study has presented a plausible problem of the catalytic effect of ClO_2 DBPs with coagulant ion by mainly focusing on chemical formation under metal ion (Cu(II) and Fe(II)) [20]. Other previous studies are mainly focused on chemical modification of the membrane or ClO_2 exposure condition by simulating soaking membrane coupon into a static and highly concentrated ClO_2 solution (10 ppm and 20 ppm, respectively) [21,22]. However, in a real RO process, membranes are pressurized by feed solution

* Corresponding author. Fax: +82 31 290 7549.
E-mail address: amjang@skku.edu (A. Jang).

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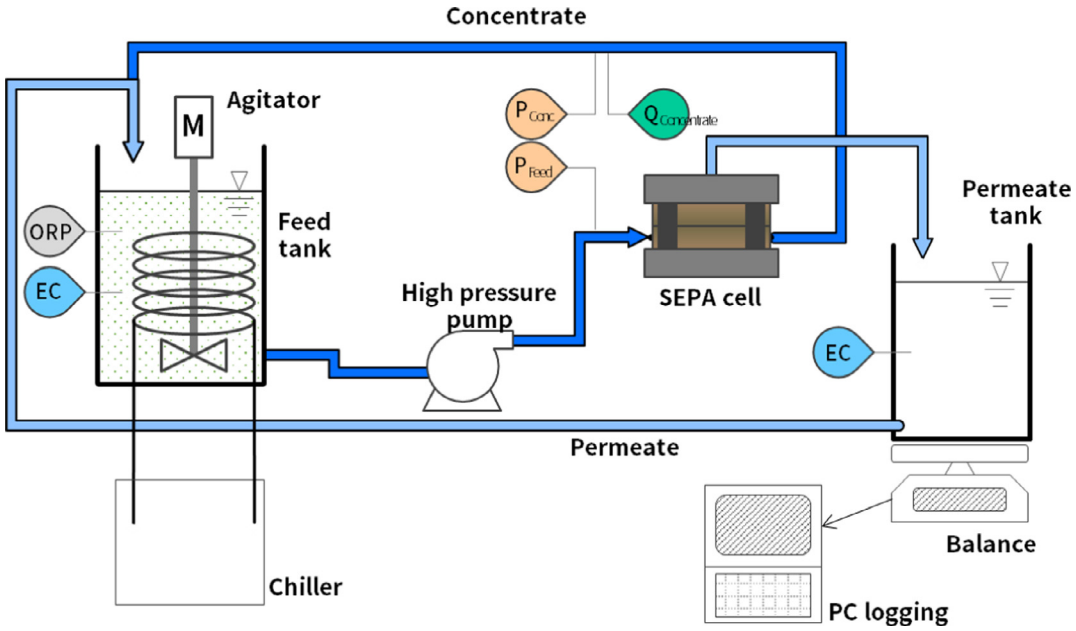

Fig. 1. Lab-scale RO membrane cell system (P: pressure gauge; Q: flow meter; EC: electrical conductivity; and ORP: oxidation reduction potential meter).

and the concentration of the disinfectant does not exceed 1.5 ppm-Cl_2. Therefore, results from previous studies are somewhat limited. In this study, the effects of two different disinfectants (NaOCl as available Cl_2, and ClO_2) on seawater RO system were examined in respect to RO performance and chemical deformation of the membrane. Additionally, a reductant was applied to the disinfectant exposed membrane to check performance recovery.

2. Materials and methods

2.1. Lab scale RO membrane cell system

A schematic diagram of a lab-scale RO membrane system is shown in Fig. 1. Satisfying general operating requirements in ASTM D4194-03, this system includes a heat exchanger for maintaining constant water temperature, an accumulator (damper) for steady flow circulation, and a back pressure (bypass) valve [23]. The feed water goes through the pipe of marine grade 316 stainless steel and undergoes pressurizing with high pressure pump (D/G-03-S, Hydra-cell, USA). It goes into a SEPA membrane cell (Siho EnC, Korea) whose cross-sectional flow area is 128.35 cm^2. It is separated into concentrate and permeate. The concentrate flows back to the feed tank while the permeate water is stored at permeate tank. The weight was measured on an electric balance (FX-3000i, AnD, USA). The electrical conductivity (EC), oxidation–reduction potential (ORP), and pH of the feed water were measured simultaneously during experiments. All electrodes used in the experiment including EC electrode (Orion 013005MD, Thermo Electron Corp., USA), ORP electrode (Orion 9180BNM, Thermo Electron Corp., USA), and pH electrode (Orion 8156BNUWP, Thermo Electron Corp., USA) were calibrated at least once a week. Once they were soaked into the feed water tank, they were retained at their initial position without moving to obtain stable reading. The ORP meter could measure redox potential as an index of disinfection potential [24,25]. Since the ORP meter had delicate reading sensitivity, its maintenance was carefully conducted by soaking the electrode in 0.1 M hydrochloric acid. The 0.1 M hydrochloric acid was substituted with fresh solution once a week. Water flux and salt rejection were calculated using data from the electrical balance and EC meter using the following equations (Eqs. (1) and (2), respectivley). Both permeate conductivity and ORP were measured when the volume reached 1000 ml.

$$Water\ flux\ \left(l * m^{-2} * hour^{-1},\ LMH\right) = \frac{Permeate\ volume}{membrane\ area\ \times\ time} \tag{1}$$

$$Salt\ rejection\ (\%) = \left(1 - \frac{conductivity\ of\ the\ permeate}{average\ of\ the\ initial,\ final\ feed\ conductivity}\right) \times 100 \tag{2}$$

2.2. Preparation of feed solution

Previous studies were conducted mainly with certain concentration of sodium chloride (NaCl) solution for membrane oxidizing experiment. In this study, synthetic seawater was prepared to simulate multi-ion effect of seawater to generate intrinsic buffer intensity. In some cases, synthetic seawater has certain Cl_2 demand that is much higher than that of deionized water or NaCl solution in 3.5% salinity [26]. Therefore, the performance results with synthetic seawater would be more applicable to real desalination process than NaCl solution.

Fifteen liters of synthetic seawater were prepared following the ASTM D1141-98 [27] right before each experimental cycle. Depending on the recipe in the ASTM (Table 2), Stock 1 and Stock 2 were prepared and stored at 4 °C without any light exposure. The following solutions were mixed together in a feed tank in a total volume of 15 l : 371.73 g of NaCl (99% NaCl, Samchun), 62.35 g of sodium sulfate (98.5% Na_2SO_4, Samchun), 300 ml of Stock 1, and 150 mL of Stock 2. The mixture was agitated for 30 min followed by 1 hour of pipe circulation with 4 liters per minute (LPM) flux without pressure to dissolve each reagent completely. The initial condition of this synthetic seawater showed certain characteristics (Table 1). Osmotic pressure was calculated using the following Eq. (3) addressed in ASTM D4516 [28]. Results are shown in Table 2. Note that the initial ORP value under 300 mV was considered as "zero" because dissolved oxygen acted as a weak oxidizer in the DI

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