

Contents lists available at ScienceDirect

Journal of Industrial and Engineering Chemistry

journal homepage: www.elsevier.com/locate/jiec

Formation and speciation of haloacetic acids in seawater desalination using chlorine dioxide as disinfectant



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ARTICLE INFO

Article history: Received 13 April 2014 Received in revised form 4 September 2014 Accepted 28 October 2014 Available online 12 December 2014

Keywords: Seawater desalination Chlorine dioxide Disinfection byproducts Haloacetic acids

ABSTRACT

To evaluate the formation potential of disinfection byproducts (DBPs) in a SWRO system when chlorine dioxide (ClO₂) is used as a chemical disinfectant, the impact of principle precursors such as natural organic matter (NOM) and bromide ion on haloacetic acids (HAAs) formation and its speciation was investigated. In the presence of high bromide levels found in seawater, the active oxidizing agents formed by the action of ClO₂ can react with NOM, resulting in halogenation reactions, and the production of brominated DBPs as the predominant byproducts. Elevating the initial levels of organic substances and bromide ions not only enhanced the total content of HAA₅ formed, but also shifted the speciation of HAAs from the chlorinated to fully brominated forms at higher disinfectant doses and longer contact times of ClO₂ disinfection. In addition, HAA₅ formation potential increased in the order of transphilic > hydrophobic > hydrophilic NOM, suggesting HAA₅ yield formed during ClO₂ disinfection significantly depends on the characteristics of NOM that can lead to different speciation distribution of DBPs. However, at high doses of ClO₂, it was observed that the disinfectant dose plays a more important role in speciation distribution of HAA₅ than the characteristics of organic precursors.

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Introduction

Seawater reverse osmosis (SWRO) is a promising desalination technology for providing drinking water as a sustainable alternative source of freshwater, which can alleviate water scarcity. The performance and safety of a SWRO system depends on the design, management and operation of four principal factors: feedwater as an intake source, treatment (pre-treatment, SWRO process and post-treatment of permeate), distribution of product drinking water, and disposal of brine. In fact, most RO system failures usually result from an inadequate pretreatment system and even require a higher degree of pretreatment as compared to thermal desalination processes in order to achieve a low content of suspended solids and silt in the water. This is because the SWRO membranes are not designed to handle high loads of filterable dissolved materials that can cause reduction of membrane life and

* Corresponding author. Tel.: +82 31 290 7526; fax: +82 31 290 7549. *E-mail address:* amjang@skku.edu (A. Jang). deterioration of product water quality [1]. It is of critical importance that disinfection improves the quality of water by controlling microbial growth, which ensures an optimal performance in desalination plants for both pretreatment and water quality after desalination.

Disinfectants are applied during pretreatment to both remove some contaminants and control microbial growth resulting in the protection of membranes from biofouling and a subsequent rapid loss of membrane integrity [1,2]. Post-treatment of permeate produced by the desalination system is needed for two key reasons prior to final distribution and use: (1) to protect public health by blending it with source water for mineralization and by post-disinfection eliminating pathogens; and (2) to make the permeate compatible with the water distribution system by chemical conditioning which increases alkalinity and stabilizes the product water.

Chemical chlorine disinfectants such as chlorine, chloramine and chlorine dioxide have all been used as disinfection and oxidizing agents for water treatment since they can effectively control excessive microbial growth in the intake, pretreatment, RO

http://dx.doi.org/10.1016/j.jiec.2014.10.046

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process and distribution system. However, they can also lead to the formation of undesired halogenated disinfection by-products (DBPs) with potential health concerns, by reacting with natural organic matter (NOM) and the halogen species (chloride, bromide or iodide) in water, all of which have been identified as the principal precursors [3]. In SWRO plants, many of the halogenated-DBPs formed during pre-treatment are removed by the desalination process, but are subsequently discharged in the brine where they can affect downstream aquatic ecosystems near outfall discharge. There they can act as herbicides and pesticides [4] if they are not diluted sufficiently prior to disposal [2]. Although the desalination process removes significant amounts of DBPs formed by peroxidation along with the other contaminants that are present [1], there are still public health concerns related to the toxic DBPs that are not completely eliminated.

Generally the organic content of desalinated water is very low, and consequently, DBPs from post-disinfection would also be expected to be low. However, this can be significantly affected by the type of blending water that is used to stabilize the water during post-treatment, especially if the source water has high organic levels. In addition, it is important to consider the amount of brominated organic byproducts that could be formed from predisinfection of seawater containing bromide, and from disinfection of blending waters, if bromide is reintroduced to the finished waters [5]. This is a concern since bromide (Br⁻), the anion of element bromine (Br), is generally present in seawater in relatively large amounts (65 to 155 mg/L) as compared to freshwater (<0.5 mg/L) [6,7]. Thus even high removal efficiency (e.g. > 95%) still allows some bromide, on the order of 1 to several mg/L, to be present in the finished water [8]. The DBPs produced from post-disinfection with blending water that contain additional DBP precursors such as organic substances and bromide ions are also important issues to be considered as well as the type of post-disinfection and the length and conditions of distribution system.

Although chlorination is the most common disinfectant used in current desalination systems, chlorine dioxide (ClO₂) has gained popularity for disinfection of seawater desalination as an alternative chemical disinfectant. In practice, several large scale desalination plants including the facilities in Tampa Bay, Florida, Spain and UAE increasingly employ chlorine dioxide for pre- and post-treatment disinfection [2]. Chlorine dioxide is used to provide a CT disinfection credit to inactivate pathogenic microbes, and as a preoxidant to control taste and odor compounds and other contaminants. It has been reported that chlorine dioxide is an alternative to chlorine because it is an oxidizing agent rather than a chlorinating agent, and therefore, will not form as many chlorinated DBPs, such as trihalomethanes (THMs) and haloacetic acids (HAAs), that are the two major classes of DBPs found under typical water treatment conditions [9,10].

Even though HAAs (monochloroacetic acid (MCAA), monobromoacetic acid (MBAA), dichloroacetic acid (DCAA), bromochloroacetic acid (BCAA), dibromoacetic acid (DBAA), bromodichloroacetic acid (BDCAA), dibromochloroacetic acid (DBCAA), trichloroacetic acid (TCAA), and tribromoacetic acid (TBAA)) containing chlorine and bromine can be produced from chlorination of waters in the presence of bromide, only five of them, called as HAA₅ (MCAA, DCAA, TCAA, MBAA, and DBAA), are regulated by the US EPA under the Disinfectants/Disinfection Byproducts (D/DBP) Rule due to limited formation and occurrence data for some of the species [11]. Due to their carcinogenic health risks, the DBP regulation requires utilities to comply with the maximum contaminant level (MCL) of HAA₅ of 60 μ g/L for the running annual average at individual locations in a distribution system [11], which requires various treatment and disinfection strategies [12].

Although much of the research on DBPs formation by ClO_2 has focused on freshwater [13–16], there are still a limited number of available data on DBP formation in desalination systems, and many of the published studies were conducted at distillation plants [2]. In addition, the formation of DBPs from ClO_2 in an SWRO has not been reported even with the increased popularity of ClO_2 as a promising chemical disinfectant for desalination systems. However, research on the use and effect of ClO_2 for pretreatment, on the level and type of DBPs in the desalinated seawater produced by SWRO systems needs to be evaluated [1].

In this study, an effort has been made to investigate the effect of organic precursors and bromide ion in seawater (feed to desalination plants) on the formation potential of HAA₅ as well as the effects of ClO₂ dose and disinfection contact time. This study was designed to simulate the formation of chlorinated and brominated organic byproducts when ClO₂ disinfectant is used in SWRO system for pre-treatment and for disinfection of water after desalination. The ClO₂ disinfectant is usually applied to seawater containing bromide during pre-treatment to control biofouling on intake structure and RO membrane, and improve the performance of filtration. In addition, the higher disinfectant dose occasionally required such as after a rainstorm also needs to be considered. Disinfectants also are applied prior to sending the finished water into the distribution system. Although much of the HAAs formed during pre-treatment can be removed efficiently by the RO system [17], DBPs may be formed when desalinated water is blended with water from organic matter-rich sources prior to disinfection, or when they are mixed in the presence of a residual disinfectant in the distribution system [2]. The type and level of blending water that is used post treatment to stabilize the water is a significant consideration as a source of additional DBP precursors, if organic matter and bromide are reintroduced to the finished waters. To demonstrate the possible scenarios in a SWRO plant, this study was designed to determine the impact of organic matter (concentration and different fractions) and bromide ions in seawater as the principle precursors of DBPs on the HAA₅ formation, and its speciation during ClO₂ disinfection, with a special focus on kinetic experiments using various disinfectant doses as a function of time.

Material and methods

Sources of seawater

Natural seawater was collected from the intake of a SWRO testbed plant located on Gijang beach in the city of Busan, South Korea (35°13′40″N, 129°14′20″E) (Fig. S1). The water characteristics of natural seawater collected are summarized in Table 1.

Humic acid (Sigma-Aldrich, 1415-93-6) was spiked as the source of organic carbon in the natural seawater sample to investigate the effect of NOM on the formation of HAA₅ by mimicking the organic precursors found in aquatic environment. The NOM stock solution was prepared by dissolving 1 g of humic acid in 20 mL of 0.1 N NaOH, adjusting the pH to 8, and filtering through a 0.45 μ m-membrane. The stock solution was then

Table	1		
Water	characteristics	of natural	seawater.

General parameter	pН	8.1	TSS (mg/L)	7.8
	Turbidity (NTU)	3.5	TDS (mg/L)	35,780
Organic parameter	DOC (mg/L)	4.6	SUVA (L/mg/m)	1.19
Inorganic parameter	Na ⁺ (mg/L)	10,995	Mg ²⁺ (mg/L)	1,371
	Ca ²⁺ (mg/L)	435	K^{+} (mg/L)	433
	NH_4^+ (mg/L)	0.06	Cl ⁻ (mg/L)	18,795
	SO_4^{2-} (mg/L)	2,698	HCO_3^- (mg/L)	122
	$Br^{-}(mg/L)$	60	NO_3^- (mg/L)	0.05

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