Contents lists available at ScienceDirect

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

Assessment of hardness, microorganism and organic matter removal from seawater by electrocoagulation as a pretreatment of desalination by reverse osmosis

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HIGHLIGHTS

• Electrocoagulation was applied as a pretreatment for seawater desalination.

• Electrocoagulation is cost-effective due to the high conductivity of seawater.

• Natural organic matter and microorganisms were efficiently removed.

• Electrocoagulation can be used for disinfection, but not for softening.

ARTICLE INFO

Article history: Received 1 September 2015 Received in revised form 28 December 2015 Accepted 29 December 2015 Available online 8 January 2016

Keywords: Electrocoagulation Reverse osmosis Pretreatment Seawater desalination Membrane fouling Organic matter Microorganisms Total hardness Disinfection

ABSTRACT

Electrocoagulation (EC) process was used as an alternative pretreatment in order to assess its applicability to replace the conventional pretreatments used to mitigate membrane fouling prior to seawater desalination by reverse osmosis process, such as chemical coagulation, chlorination and scale inhibitors. Electrocoagulation was conducted in a batch cell using aluminum electrodes driven in the galvanostatic mode. Absorbance (UV_{254nm}) and DOC were measured to follow the effectiveness of EC process for removing organic matter from seawater. Likewise, cultivable heterotrophic bacteria were enumerated to assess the disinfection ability of the process and total hardness was monitored. The influence of various operating parameters, namely mixing speed, current density, initial pH and inter-electrode distance, was investigated. The removal of organic matter from seawater by electrocoagulation was improved with higher current density and lower pH. Electrocoagulation removed 70.8% DOC, while absorbance abatement was 89.7% with a complete removal of microorganisms at high current density. Conversely, the abatement of total hardness was weak, about 10%, so that electrocoagulation could not be used as a softening process. Finally, experimental results showed the high potential of electrocoagulation as a pretreatment method to mitigate potential organic fouling and biofouling of reverse osmosis membrane owing to its ability to remove effectively dissolved organic matter and microorganisms from seawater.

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1. Introduction

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Given the global population growth estimated between 40 and 50% in the upcoming 50 years and the growing anthropic activities, such as deforestation, industrialization and the resulting global warming, the efficient management of fresh water resources and wastewater treatment facilities for water reuse are no longer a satisfactory solution to meet the growing fresh water demand on the long term [1]. 96.5% of

Earth water being seawater in comparison to 0.8% for fresh water, desalination of seawater seems to be the most appropriate answer to overcome the fresh water scarcity [2–3]. Among the technologies of desalination of seawater, reverse osmosis has been recognized to be the most cost-efficient technology in comparison to thermal processes thanks to the remarkable advances in reverse osmosis membrane technology [4] and the use of energy recovery system [3]. However, the desalination industry encounters a major challenge that consists in reverse osmosis membrane fouling, which implies a higher treatment cost due to the important frequency of membrane cleaning or/and replacement [5]. Efficient seawater desalination requires a suitable pretreatment

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prior to reverse osmosis [1,6], but the conventional methods still exhibit some limits and/or drawbacks. Among them, one can cite the difficulty of chemical coagulation to achieve the removal of total organic carbon from seawater at the guideline value of $0.5 \text{ mg} \cdot \text{L}^{-1}$ below which biofouling is unlikely to occur, the reaction of chlorine with organic additives that could lead to the production of carcinogenic substances due to chlorination, and the use of scale inhibitors that can enhance the risk of membrane biofouling, as polyphosphate-based anti-scalants can serve as a phosphorous source of nutrients. Hence, recent research focuses on more efficient alternative pretreatments.

In detail, the types of membrane fouling in reverse osmosis can be split into scaling, colloidal fouling, organic fouling and biofouling. Colloidal fouling is due to the deposit of inorganic or organic colloids on the membrane, scaling is caused by the precipitation of mineral components on the membrane surface, organic fouling occurs due to dissolved organic compounds, such as humic acids and fulvic acids from the feed seawater adsorbed on the membrane, whereas biofouling takes place when there are microorganisms present in the feed seawater that adhere on the membrane [5]. Being the most harmful fouling mechanism, organic fouling should be avoided or reduced as far as possible, as the dissolved organic compounds in the feed seawater serve as nutrients to the microorganisms whose metabolism and growth could lead to severe biofouling by forming a biofilm on the membrane [7]. A small fraction of biofilm is composed of metabolites which in turn include extracellular polymeric substances that adhere on membrane, resulting on long-term irreversible fouling [8]. The latter is insensitive to physical and chemical cleanings and, as a consequence, membrane replacement is required. Biofouling is considered as the Achilles' heel of membrane processes as, even with a 99% removal of microorganisms, the remaining microorganisms can grow in the presence of the dissolved organic compounds and lead to membrane biofouling [9]. Thus, alongside a disinfection process, an efficient process to remove organic matter is highly needed to avoid or slow down membrane biofouling.

Conventional pretreatment technologies (such as clarification/ dissolved air flotation, coagulation-flocculation, scale inhibitors, sedimentation and granular media filtration) and non-conventional pretreatments (such as membrane processes including microfiltration, ultrafiltration) are efficient to avoid membrane fouling, except organic fouling and bio-fouling [1,10]. The natural organic matter is removed from seawater by two main conventional physicochemical processes, namely chemical coagulation using FeCl₃ followed sometimes by flocculation [6,11], and adsorption using powdered activated carbon [1], while chlorination is commonly used to prevent biofouling by killing or impairing the viability of microorganisms [5,12]. Hybrid processes, namely coagulation coupled with microfiltration or ultrafiltration and adsorption coupled with microfiltration or ultrafiltration, have been used as pretreatment methods prior to seawater desalination by reverse osmosis in attempt to reduce considerably the dissolved organic matter [13]. However, the results are not very convincing, as an important fraction of organic matter in seawater corresponds to low molecular weight dissolved organic molecules [14].

Having proven its high efficiency for the abatement of a wide range of pollutants [15], electrocoagulation is gaining an increasing interest as a promising alternative pretreatment method prior to membrane processes, chiefly microfiltration [16–17], ultrafiltration [18] and nanofiltration [19–20] in water treatment. Electrocoagulation turned out to be an efficient pretreatment prior reverse osmosis to remove silica from brackish water [21]. Timmes et al. [22] investigated the pretreatment of seawater by iron electrocoagulation prior to ultrafiltration; as a result, electrocoagulation turned out to be a competitive and feasible pretreatment that reduced the transmembrane pressure and hydraulic resistance. In comparison with chemical coagulation, the hydraulic and chemical cleaning of ultrafiltration membrane fouled with the seawater pretreated by electrocoagulation showed higher water flux recovery and lower hydraulic resistance: it was finally concluded that electrocoagulation pretreatment may extend the life of ultrafiltration membrane [22].

However, studies on the pretreatment of seawater by electrocoagulation are almost non-existent even if electrocoagulation process has been successfully applied to remove the same kinds of pollutants from wastewater/water: namely, turbidity, colloids and particulate compounds [23-24], organic matter [25-27], hardness [23,28] and microorganisms [29] that are responsible for all types of reverse osmosis membrane fouling in seawater desalination. In addition, in comparison with conventional chemical coagulation, electrocoagulation as a pretreatment prior to the seawater desalination by reverse osmosis may provide additional advantages, such as the possibility to oxidize chloride anions Cl⁻ in active chlorine that could act as an oxidizer of organic compounds or could play the role of a disinfectant for the microorganisms. Furthermore, the high conductivity of seawater could result in the reduction of energy consumption and in a high faradaic efficiency of anode dissolution. The electric field generated by the application of electric current between electrodes during electrocoagulation may also contribute to the disinfection of seawater through cell disruption or inactivation that results from its effect on cell membrane permeability. On the basis of recent works on the removal of natural organic matter from water by electrocoagulation process, the comparison of electrode materials has shown that a better removal efficiency was obtained with aluminum electrodes than iron electrodes and hybrid electrodes (composed of aluminum and iron) [27,30]; so, aluminum electrodes were chosen for this study.

The present work aims at assessing and validating the applicability of electrocoagulation as an alternative pretreatment prior the seawater desalination by reverse osmosis in order to prevent or to mitigate significantly the reverse osmosis membrane fouling, especially biofouling and organic fouling. An in-depth study was conducted by following the abatement of the compounds and species that are responsible for fouling, such as seawater organic matter, microorganisms and hardness for organic fouling, biofouling and scaling, respectively, throughout electrocoagulation pretreatment in the batch mode. The objective is to estimate the efficiency and the limits of the electrocoagulation pretreatment.

2. A brief description of electrocoagulation

Electrocoagulation is an electrochemical process whereby metal cations are generated in situ from the electrodissolution of sacrificial anode under an action of electric current applied between the electrodes. These metal cations are prone to hydrolysis, leading thereby to the metal hydroxide species, depending on the pH of aqueous medium, which will behave as coagulants and/or adsorbents and/or an electrostatic attractor of pollutants for their removal. The oxidation reaction takes place at the anode and the reduction of water at cathode. Thus, the respective electrochemical reactions are summarized as follows:

• At the anode : $Al \rightarrow Al^{3+} + 3e^{-}$ (1)

• At the cathode :
$$2H_2O + 2e^- \rightarrow H_2 + 2OH^-$$
 (2)

Alongside the aforementioned reaction at the anode, secondary reactions may occur in case of high anode potential, such as oxidation of Cl⁻ ions present in water into chlorine that is in turn transformed in ClOH/ClO⁻ through oxidation by water. Unlikely to the oxidation of water in O₂ that lowers the faradaic efficiency of Al release, the oxidation of Cl⁻ in Cl₂ and ClOH which are strong oxidizers may be beneficial for water treatment. It is worth noting that a higher anode potential could also lead to a direct oxidation of organic compounds.

$$2Cl^{-} \rightarrow Cl_{2} + 2e^{-} \tag{3}$$

$$Cl_2 + H_2 O \rightarrow ClOH + Cl^- + H^+ \tag{4}$$

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