



# Study of fouling and scaling in capacitive deionisation by using dissolved organic and inorganic salts

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## HIGHLIGHTS

- ▶ Dissolved organic matter (humic acid) is the main cause of CDI electrode fouling.
- ▶ CDI removal efficiency and its production rate decreases by increasing feed TOC.
- ▶ CDI energy consumption ( $\text{kWh/m}^3$ ) increases by increasing feed TOC.
- ▶ Ferric ions seemed to be a great contributor to CDI scaling.
- ▶ Alkaline and acid cleaning solutions were able to completely restore the recovery of the CDI.

## ARTICLE INFO

### Article history:

Received 1 August 2012  
Received in revised form  
28 November 2012  
Accepted 28 November 2012  
Available online 7 December 2012

### Keywords:

Capacitive deionization  
Fouling  
Total organic carbon  
Energy consumption  
Humic acid

## ABSTRACT

In this work, fouling, scaling and cleaning of the capacitive deionisation (CDI) with activated carbon electrodes were systematically investigated for the first time. Electrode fouling caused by dissolved organic matter using sodium salt of humic acid as a model foulant (measured by total organic carbon concentration, TOC) and inorganic salt ( $\text{NaCl}$ ,  $\text{MgCl}_2$ ,  $\text{CaCl}_2$  and  $\text{FeCl}_3$ ) in the CDI feed solutions was investigated in a series of controlled fouling experiments. After each CDI experiment, a series of cleaning steps was performed to understand the reversibility of fouling accumulated on the electrode surface by analysing the cleaning solutions. The higher the TOC concentration in the CDI feed solution, the more the reduction of salt removal efficiency, declination in the production rate and energy consumption. Dissolved organic matter is the main cause of electrode fouling, as it blocks the activated carbon pores and reduces their electrosorption capacitance. Ca and Mg have no noticeable effect on the CDI treatment performance. However, Fe seemed to have a greater effect on CDI electrode fouling. Alkaline and acid cleaning solutions were able to restore the recovery of the CDI performance from fouling. Pre-treatment to reduce the dissolved organic matter levels is recommended to achieve sustainable treatment performance.

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## 1. Introduction

Water shortage has become one of the major problems in the world. The global water crisis has drawn much world attention on finding sustainable ways to produce fresh drinking water. Many researchers have indicated that water will be the oil of the 21st century [1–3]. Scarcity of fresh water may lead to warfare, as oil did in the past. About 98 per cent of the planet's water is either sea water or brackish water. Therefore, we have to explore novel, more efficient and energy effective ways to purify salty water. Currently, most of the water desalination technologies are energy and cost intensive and can cause secondary pollution [4].

Capacitive deionisation (CDI) technology stands out as one of the most useful tools for brackish water desalination. CDI is an

energy-efficient desalination technology because it operates at a relatively low electrical voltage (typically 0.8–2.0 V) at which no electrolysis reaction occurs [5,6]. Other advantages of using CDI technology include no need for pressure-driven membranes or high-pressure pumps, simplicity of design, low cost, and operation at ambient conditions [7–9]. The CDI energy consumption is about three times lower than that of the electrodialysis reversal process [10]. The production cost of CDI is about three times lower than that of the low-pressure RO process [11]. The CDI process cycle consists of two phases; namely, purification and regeneration. During the purification phase, an electric potential is applied to electrodes, which forces charged ions in the electrolyte solution to migrate towards oppositely charged electrodes, to be held in the electrical double layer. Regeneration then takes place by reversing the applied potential to expel the adsorbed ions into the waste stream [12,13].

Although much research has been conducted on CDI, issues such as fouling and scaling remain to be more thoroughly

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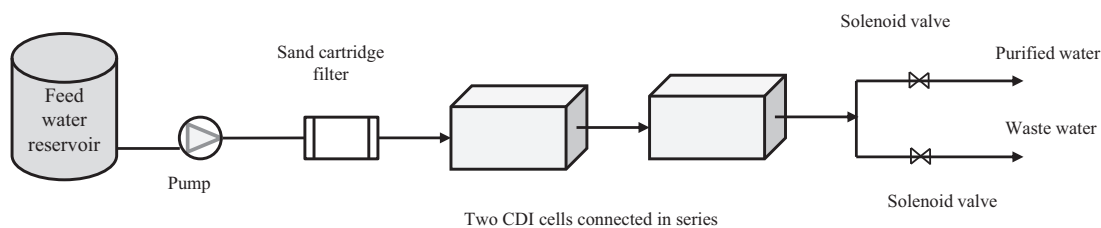


Fig. 1. Schematic diagram of the CDI pilot plant unit.

investigated before CDI can be widely accepted in applications. Most research efforts have focused on developing novel electrode materials and enhancing their properties [14–17]. CDI technology has also been used to remove inorganic salts from different types of water [7,8,11,18–21]. Until now, there has been a dearth of research reporting the effect of dissolved organic matter in brackish water on CDI operation performance and treated water quality. Humic acid is one of the dissolved organic matter which is abundant in groundwater, natural water and wastewater [22–24]. Humic acid is widely used as a typical model compound to form organic fouling layers on membrane or other materials by many membrane filtration researchers [24–26]. No previous study has investigated the organic fouling of CDI electrodes and the effect of this on CDI performance. The scaling problems commonly associated with membrane technologies may be avoided using CDI [7,12]. The most common scalants for reverse osmosis technology are Ca, Mg and Fe [27,28]. CDI scaling and fouling may result in a decline in the quality of purified water and a reduced production rate [5]. Therefore, it is necessary to study the effect of dissolved organic and inorganic compounds on the treatment efficiency of CDI caused by the accumulation of foulants on the activated carbon electrodes. Further, it is essential to identify effective cleaning methods for the electrodes, to remove fouling caused by different types of foulants. Such an improvement in the understanding of fouling issues for CDI's is very timely, to employ CDI as a practical desalination technology in fresh water augmentation.

The objectives of this study are to understand the effect of the different foulants in feed solution on CDI activated carbon electrodes and to identify the composition of the foulants accumulated on the surface of the electrodes during the treatment of brackish water. In this study, investigation was conducted using a prototype CDI unit with the capacity of 1000–2000 L/day at laboratory. To investigate the effective cleaning method for CDI electrodes, a series of specific cleaning protocols was conducted. The used cleaning solutions in each cleaning step were collected and analysed to identify the major foulants of the CDI electrodes. The effect of CDI fouling on the treatment efficiency, process flow rate and energy consumption was determined as well. This investigation provides valuable information on operation and maintenance regarding potential fouling of the CDI.

## 2. Experimental

### 2.1. The CDI pilot plant

Fig. 1 shows the combined pretreatment-CDI unit used in this study. The CDI cells (Aqua EWP, USA) were comprised of porous activated carbon electrodes with a specific area of  $800 \text{ m}^2/\text{g}$ ; a flow rate of  $2 \text{ L}/\text{min}$  was used during all the experiments. The operating direct current voltage of the CDI process was  $1.5 \text{ V}$ . The process cycle was controlled by a programmable logic controller. Each cycle consisted of a purification stage lasting  $1.5 \text{ min}$ , and a regeneration stage lasting  $1 \text{ min}$ . The regeneration step commences with  $30 \text{ s}$  of flushing, when the effluent valve and the influent valve are closed

and the power supply is off, followed by another  $30 \text{ s}$  of flushing when the effluent waste valve and the influent valve are opened and the power is turned on with the opposite polarity of  $1.5 \text{ VDC}$ . In the purification stage, the influent valve and the effluent purification valve are opened and the power is turned on with the polarity of  $1.5 \text{ VDC}$ . The CDI unit included two electrode cells connected in series (Fig. 1). Each cell assembly contained 100 pairs of activated carbon electrodes with dimensions of  $158 \text{ mm} \times 174 \text{ mm} \times 0.3 \text{ mm}$  and a total mass of 1354 grams of activated carbon. The CDI prototype unit and the electrode material is reviewed in more detail and extended in Refs. [29,30]. The fouling and scaling of the CDI cells were assessed by observing the decrease in permeate flow rate and electrosorption removal efficiency during the purification cycle, and by comparing the CDI operational performance using various feed solution compositions. For each feed composition, the CDI unit was operated continuously for  $30 \text{ h}$  using various feed solutions followed by as series of cleanings steps.

### 2.2. Experimental methods

A series of laboratory experiments was conducted to investigate the effect of feed solution composition on CDI performance; that is, the permeates flow rate, electrosorption removal efficiency and electrical energy consumption. Three different feed solutions (F1, F2 and F3), as shown in Table 1, were used to identify the organic fouling potential of the CDI, while other two feed solutions (F4 and F5) were used to investigate the scaling potential of the unit. F6 solution was used to investigate the effect of both organic and inorganic salts together on the CDI operational performance. The chemicals used to prepare the CDI feed solutions with various compositions were sodium chloride ( $\text{NaCl}$ ), calcium chloride ( $\text{CaCl}_2$ ), magnesium chloride ( $\text{MgCl}_2$ ), ferric chloride ( $\text{FeCl}_3$ ) and sodium salt of humic acid. In this study, humic acid sodium salt as a model compound of dissolved organic matter was added in the synthetic feed solution. In order to determine the concentration of the dissolved organics in the treated water, instead of using other analytical method, such as HPLC, total organic carbon (TOC) analyser was used to measure the concentration of dissolved organics. Sodium salt humic acid was obtained from Sigma–Aldrich. Sodium salt humic acid has an average molecular weight ranged from 4000 to 23,000 [31]. The total acidity is  $5 \text{ m mol}/\text{g}$ , with a carboxylic acidity value of  $3.4 \text{ m mol}/\text{g}$  [24]. The solution chemistry of feed solution of humic acid is described in more detail and in Refs. [24,31,32]. The pH of all the feed solutions was in the range of 6.7–7.1, which is close to groundwater pH in previous studies [30,33]. No pH adjustment for the feed synthetic solution was carried out through the whole study. The effect of organic fouling and scaling of CDI electrodes was reflected as a declining of flow rate and salt removal efficiency. The flow rate of the CDI unit was continuously monitored throughout the whole experiment period. Samples of both purified and waste stream were analysed periodically. The conductivity and total dissolved solids (TDS) were measured using an electrical conductivity metre (HACH, HQ40d digital metre). pH values were measured using a PHM201 device. A Cary 100 Bio ultraviolet

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