



Improved sea water quality by removal of the total hardness using static step-by-step deposition and extraction technique as an efficient pretreatment method



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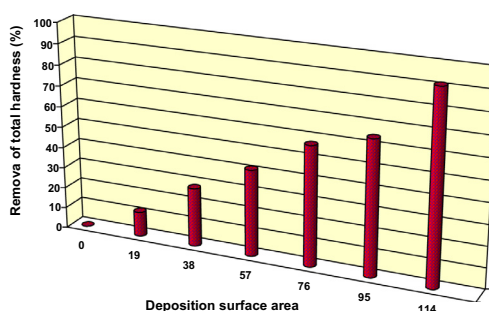
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HIGHLIGHTS

- A pretreatment method of sea water from total hardness and scale formation.
- Over 91.5% removal of total hardness by using static step-by-step deposition and extraction technique.
- Several experimental controlling factors were evaluated.
- The number of dipping cycles and exposed surface area are highly significant.
- The pH value of sea water dropped from 8.0 to 6.5.

GRAPHICAL ABSTRACT



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ABSTRACT

The static step-by-step deposition and extraction technique (SbS-DET) was explored as a prior methodology for efficient and low cost treatment of total hardness in sea water. Removal of Ca(II) and Mg(II) ions from sea water samples was based on sequential surface deposition in the form of 8-hydroxyquinolate complexes. Several important experimental controlling factors were examined, optimized and evaluated in this study such as the effect of dipping time, reaction temperature, initial concentration of 8-hydroxyquinoline ligand, number of dipping cycle and exposed surface area. The results of this study refer to an excellent and efficient removal of total hardness from the investigated sea water samples. The number of dipping cycle and exposed surface area were characterized from this study as the most important experimental controlling factors. The total hardness was dropped from 4629 to 381.9 mg L⁻¹ as CaCO₃ with a 91.75% removal value by using 30 dipping cycle, 3 min dipping time and 76 cm² surface area. The same trend was also obtained by producing a 91.47% removal value of the total hardness when the exposed surface area was used as 114 cm². The progress of SbS-DET was also characterized by a gradual drop in the pH value of sea water from 8.0 to 6.5 upon completion of the removal process.

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1. Introduction

The world is now struggling to obtain pure water supply for use in all purposes such as municipal and industrial applications. This

is mainly due to the increased population growth which is faced by a widespread shortage in water supply. Moreover, about 96% of the total global water is mainly present as sea and ocean saline water and only about 1% is generally regarded as the fresh global water [1]. By using the proper treatment methods for seawater such as desalination technologies, treated water can be beneficial in different fields such as drinking water, agricultural and industrial

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purposes [2]. Generally, desalination technology is classified according to the separation mechanism either thermal or membrane based technique. Desalination process of sea water by reverse osmosis (RO) or membrane filtration is generally suffering from a number of well known problems [3]. One of these is known as the scaling which is produced by the presence of hardness in sea water. The scaling problem leads to surface deposits due to the presence of some common species such as calcium, magnesium, iron, copper and other elements as well as silica [4]. Therefore, scaling results in precipitation of these species in the form of insoluble inorganic salts on the membrane surface. In addition, scale formation is generally associated with either a chemical or physical change in the water quality or in the distribution system. Temperature, pressure or pH change can also trigger scale formation in sea water as well as some other industrial applications such as boilers, cooling towers and similar water treatment or delivery systems [5].

The large seawater RO desalination plant consists of four major components. The initial stage in this process is mainly based on the feed water pretreatment unit [6,7]. A typical pretreatment includes precipitation [8,9], coagulation [10–12], flocculation [13], sedimentation [14], chlorination [15,16] and filtration [17] as well as ultra-filtration, microfiltration and nanofiltration [13]. The selection of a definite type of pretreatment method depends on the characteristics of feed water, configuration, recovery ratio, and quality of the product water. On the other hand, the common methodologies used for water treatment of scaling problem in various operation systems are mainly based on the minimization of water hardness by lowering the pH value of water supply, addition of sequestering agents, optimization of the operational condition, installation of water antiscalant conditioning systems and addition of water softening components. However, most of the modern interest in desalination process is mainly focused on developing low-cost pretreatment methods for providing accepted quality of water [18].

Static step-by-step deposition and extraction technique (SbS-DET), also known as layer-by-layer (LbL) chemical deposition technique, or successive ion layer adsorption and reaction (SILAR) technique [19–22], has opened new promising ways for static removal and speciation of some metal cations from aqueous and nonaqueous solutions [23–26]. This technique is now well known as a very simple, inexpensive, and rapid procedure as well as a highly versatile approach for fabrication of controlled layered structures from various kinds of component materials. The high versatility and flexibility of this assembly is recently implemented in various disciplines to generate new concepts, new procedures, new materials, and new applications [27]. The fundamental principle of this technique is basically dependent on the selection of appropriate interacting deposition species which can react and deposit the target compound on the surface of solid substrates. In the case of metal cations, a suitable ligand must be selected for the formation of the target deposited complex materials without any possibility of back dissolution in the aqueous or nonaqueous matrices [25,26].

The purpose of this work is to explore the feasibility of static step-by-step deposition and extraction technique (SbS-DET) as a potentially high performance, low cost and non-energy consumption pretreatment step for removal of the total hardness and prevention of scale formation in natural seawater samples. The effectiveness of this process in removal of dissolved cations such as calcium and magnesium was investigated and optimized under variable conditions of dipping time, reaction temperature, ligand concentration, number of dipping cycle and exposed surface area. 8-Hydroxyquinoline ligand was used due to its excellent tendency to form metal complexes with Mg(II) and Ca(II). In this work, we also followed and monitored the change in the real pH value of sea water as a measure of the total hardness removal during the progress of SbS-DET.

2. Experimental

2.1. Chemicals and materials

Analytical grade 8-hydroxyquinoline (8HOQ), ethyl alcohol, hydrochloric acid, ammonia solution, ammonium chloride, Eriochrome Black T (3-hydroxy-4-(1-hydroxy-2-naphthylazo)-7-nitro-1-naphthalene sulfonic acid), EDTA disodium salt, sodium tetraborate, sodium hydroxide, hydroxylammonium chloride were purchased from Aldrich Chemical Company, USA and BDH Limited, Poole, England and used as received.

2.2. Instrumentations

A calibrated Metrohm 780 pH-meter against standard buffer solutions, 4.00, 7.00 and 9.00 ± 0.01 was used to measure the pH-value of the examined sea water samples. Scanning electron microscope (JSM-6360LA, JEOL Ltd.), (JSM-5300, JEOL Ltd.) and an ion sputtering coating device (JEOL-JFC-1100E) were used to examine the images of deposited materials on the glass substrate.

2.3. Sampling

Seawater samples were collected from the El-Shatby beach located on the north Mediterranean shore of Alexandria, Egypt. Ten-liter sample of seawater was collected and stored at 20 °C. Regular tests were conducted to characterize the collected samples and to ensure that the water quality remained consistent. To prevent any contamination of the collected samples, the containers were cleaned properly in the laboratory prior to sampling and rinsed at least twice with seawater at the site prior to the sample collection. Sea water sample was tested by determination of the total hardness and pH value and the averaged results of these two parameters are listed in Table 1.

2.4. General procedures of SbS-DET

The static step-by-step deposition and extraction technique (SbS-DET) for removal of total hardness from seawater samples was investigated by using a glass micro-slide of the size 76.0 mm, 25.0 mm and 1.0 mm as the solid substrate. The glass micro-slides were cleaned as previously reported [23–26]. The clean glass substrate was vertically immersed into a 50.0 mL of 0.1 M-ethanolic solution of 8HOQ for a period of 3.0 min followed by air drying for additional 2.0 min. The substrate was then immersed into a 50.0 mL of seawater sample for 3.0 min whereas the pre-deposited 8HOQ molecules on the surface of glass substrate were allowed to deposit both of Ca(II) and Mg(II) in the forms of 8-hydroxyquinolate complexes. A drying period of 2.0 min was then applied on the glass substrate and the previous procedures were denoted as a single dipping cycle. The number of dipping cycles was increased up to 30 and the final residual total hardness in sea water sample was subjected to complexometric EDTA-titration [28].

2.5. Optimization of controlling factors

Several important experimental controlling factors were examined, optimized and evaluated including the effect of (i) dipping

Table 1
Characteristics of the collected seawater used in the experimental procedure.

Parameter	Unit	Result	Standard deviation
pH	pH units	8.00	±0.10
Total hardness	mg/L as CaCO ₃	4629.00	±18.0

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