



Application of micellar-enhanced ultrafiltration in the pre-treatment of seawater for boron removal



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ARTICLE INFO

Keywords:

MEUF
Boron
Sodium dodecyl sulphate
Membrane processes
Desalination

ABSTRACT

In this research, micellar enhanced ultrafiltration (MEUF) is tested as a seawater pre-treatment before entering RO. The experimental tests were performed by means of monotubular ceramic membranes of 210 kDa and 1 kDa. Boron is removed from the water flow using sodium dodecyl sulphate as a surfactant. The synthetic solutions contain 5 mg/L of boron and SDS concentrations are equal to 1.15 g/L and 2.88 g/L, under and above the critical micellar concentration, respectively.

Experimental data showed that MEUF is efficient in removing boron, with best performances obtained at low pressure, with a low or null surfactant concentration for 1 kDa membrane and a high surfactant concentration for 210 kDa membrane. Moreover, experimental results have been used to perform a preliminary process analysis for a hypothetical sea-water desalination plant, with MEUF as a pretreatment for the RO section. The results showed that the MEUF guarantees a boron concentration after RO below the allowed threshold, by using a single step osmosis; moreover, in this way it was possible to reduce the energy consumption thus resulting in an appreciable reduction of carbon footprint as well as of the unit cost of water.

1. Introduction

Over the past decades, the number of countries experiencing water scarcity has significantly increased significantly due to the population growth and to the greater demand for water in industrial, agricultural and domestic activities [1]. Within 2025, it is estimated that 1.8 billion people will live in areas experiencing the scarcity of water [1]. Because of the growing demand for soft water and the peer decline in sources due to the anthropic pressure and the climate change, today it is mandatory to find alternative water resources [2]. In this regard, seawater desalination represents and has already become in some parts of the world, a possible and efficient method to obtain clean water [3]. On the other hand, for a safe use of desalination technologies, mainly for the membrane-based processes, it is mandatory to monitor and control concentration levels of several natural compounds such as boron, that are positive for human health at low concentrations, but can become toxic at high levels. Elemental boron, boron oxide, boric acid, borates, and many organoboron compounds are relatively nontoxic. An intake above of 4 g/day of boric acid is considered toxic and an intake above of 0.5 g/day for 50 days causes minor digestive issues and other

problems suggestive of toxicity [4]. The World Health Organization set for many years the boron concentration limit for the drinking water at 0.5 mg/L [5] (a value indeed considered a provisional guideline value since it is below the level achievable with the existent treatment) but this value was raised up to 2.4 mg/L due to the positive effect of boron on human health (at restrained levels) [6]. Anyway, the content of boron in drinking water is adjusted throughout the world. For example, in the European Union, South Korea and Japan, the maximum amount of boron in potable water is kept at the 1.0 mg/L level [7].

Until now, dissolved boron has been removed to almost zero concentrations through thermal desalination based on multi-stage flash distillation (MSF, Multi-Stage Flash) or multi-effect distillation (MED, Multi-Effect Distillation). However, thermal processes are becoming less and less implemented because of their high energy footprint in comparison to the membrane technologies [8]. The alternative approaches for boron removal can be grouped into the following categories: separation through adsorption, separation through membrane filtration and hybrid separation processes characterized by the combination of them [9].

The use of chelating ion exchange resins can be considered one of

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the most representative methods [8]. This technique can be used for seawater currents, but it is more efficient at low salt concentrations [8]. The different commercial products for boron removal, produced by different companies, are all cross-linked polystyrene macroporous resins with an active *N*-methyl-*D*-glucamine (NMG) group [10]. Their effectiveness depends on several factors such as boron concentration, flowrate, regeneration as well as on the output boron limits [10]. As reported by Li et al. [11] the presence of three or more hydroxyl groups as ligands, located in the *cis* position (the so-called “vis-diols”) engaged in the synthesis of chelating resins show a high selectivity to boron. The same groups are not reactive to other metals and elements. A new hybrid gel prepared with tetraethoxylan (TEOS), (3-glycidossiipropyl) trimethoxysilane (GPTMS), and a precursor synthesized by GPTMS and *N*-methyl-*D*-glucamine shows a good mechanical strength and affinity for boron [12]. Several studies also focused on natural polymers. Orlando et al. [13] were the first to highlight the good efficiency of natural waste materials which could be used as anion exchange resins. Pre-treated sawdust of different kinds of trees (pine, birch, oak) were used for the removal of boron, but turned out to be unfit materials [14]. A chitosan resin was obtained with NMDG using cross-linked chitosan with ethyleneglycolglycidylether (EGDE) [15]. Chitosan adsorbent with NMDG functional group, synthesized using ATRP (atom transfer radical polymerization), showed a much higher boron absorption than any commercial resin [16,17].

Membrane processes are the second group of technologies used to remove boron from water and are mostly applied for desalination of sea- and brackish water [18]. In these processes, boron rejection depends on pH values, but also on parameters such as temperature, pressure, salinity, ionic strength and suspended solids. In seawater boron is generally present as boric acid (H_3BO_3) and gives rise to a dissociation equilibrium reaction with the speciation depending on the pH of the solution. Especially at low pH, the main species is fully protonated in its neutral form; in these conditions, because of its smaller size and the lack of electrical charge, the rejection by reverse osmosis (RO) of non-ionized boric acid is low. At high pH values, instead, the dissociated form of H_3BO_3 , *i.e.* borate ion, predominates; this species in water is instead completely hydrated, thus resulting in a greater radius and a higher negative charge. This causes a higher rejection, both due to the size exclusion and the repulsion of negative membrane loads [9]. Güler et al. [19] studied the efficiency of two types of polyamide thin film composite SWRO membranes for boron removal from seawater. The results showed the positive effect that the feed seawater temperature has on the permeate flux and recovery ratio.

The same authors [20] reported their study relative to RO using conventional membrane (type FilmTec SW30-250 produced by DOW Chemical. Co) to remove boron and other substances, in particular sodium, magnesium, calcium, potassium, bicarbonate, chloride and sulphate from seawater. The results showed that a concentration of 1 mg/L of boron could be achieved under the following conditions: 55 bar of operative conditions and 11 °C of seawater temperature. Anyway, the removal efficiency for boron was lower with respect to the other considered substances, in fact a yield of about 70% was obtained for boron and more than 98% for the other compounds. Recent tests on commercial SWRO membranes (type TM820A/C/E-400 produced by Toray Industries Inc., SW30XHR-400i, SW30HRLE-400, SW30XLE-400i, SW30ULE-400i produced by DOW Chemical. Co and SWC4 +, SWC4 + B, SWC5/6 produced by Hydranautics) showed that a removal comprised between 83 and 95% could be obtained considering a pH range of 6.5–8 [21]. Lower removal efficiencies were obtained using aromatic polyamide RO membranes: 48–70% using SG UTC80 A/S and 12–30% using SG UTC80 A/S produced by Osmonics [22]. Boron rejection yields higher than 96% were obtained under a 2-pass configuration using new generation of RO membranes without pH adjustment [23].

Removal of boron was also performed using hybrid processes: a combination process of ion exchange and membrane filtration. These

systems are classified according to the solubility of the adsorbent in water. When it is soluble, the membrane filtration is performed through ultrafiltration (UF) membranes, while both UF and microfiltration (MF) are used when the sorbent is not water soluble in order to reduce the pressure of the system [24].

Regarding the hybrid methods, also Polymer Enhanced Ultrafiltration (PEUF) and Adsorption Membrane Filtration (AMF) can be found in the scientific literature. PEUF is a process in which ultrafiltration membranes reject polymer molecules and small species. A diluted permeate that can be discharged as a waste or used for a specific purpose is obtained from the process. Moreover, the PEUF produces a retentate flux with high concentration of metal ions bound to the polymer that must be separated from the polyelectrolyte in order to reuse the polymer and to reduce the operative costs. PEUF can achieve a boron reduction from 10 mg/L down to 0.4 mg/L [25]. The AMF (Adsorption Membrane Filtration) includes the adsorption of the molecules on micro-spherical sorbents, followed by microfiltration. This method was extensively studied by Kabay et al. [26–34] by using two commercially available resins with *N*-methyl-*D*-glucamine ligands (Diaion CRB 02 and BSR1) or synthesizing their own polymeric microspheres [33]. The membrane filtration module was made by polypropylene capillary membranes with a pore diameter 0.4 μm. The removal from several type of waters was checked, in particular from geothermal water [28,29], artificial solution [28–31,34] and from permeate on the first stage of SWRO treatment [27,34]. The results showed that a significant reduction of boron concentration in permeate was obtained in the first 15 min of the process. It was concluded that sorbent particle size affected significantly the economics of AMD hybrid system for water treatment. More details about these results were reported in the review of Wolska and Bryjak [7]. Other technologies were used to remove boron such as direct contact membrane distillation that involves the transfer of water vapor through a porous hydrophobic membrane in which the supply and the distillate are separated by a hydrophobic membrane [7]. The extraction with ionic liquids involves the dissolution of a water-insoluble polyhydric mixture in water by a suitable solvent which is water-immiscible and has a specific boron complexation. This technique has been specially developed for the recovery of boron from sweet brines and from output plant flowrate with boron concentrations of 1.7%. The ionic liquid *N,N*-Bis (2,3-dihydroxypropyl) octadecylamine (BPO) in 2-ethyl ethanol has proven to be very effective in liquid-liquid boric acid extraction in aqueous solutions [35].

As discussed above, boron can be removed from the water with various technologies but reverse osmosis and adsorption on resins are therefore the options that dominate the market. In the first case the complexity of the process due to the multi-step configuration and issues of scaling and fouling (related to the chemicals' consumption and to the water quality) represent the main drawbacks. In the second case, the technology is influenced by the high cost of the sorbent, and/or the difficulties in the regeneration process [36]. According to a recent analysis based on desalination as a case study, the economy promotes membranes in the case of bulk removal of boron while selective adsorbents are preferred for refining [9]. In particular, membrane technology is preferable at boron concentrations equal or above 500 mg/L [37].

The process object of the present work is the Micellar Enhanced UltraFiltration – MEUF. It includes the addition of a surfactant to the solution to be filtered at a concentration above the critical micellar concentration (CMC). When the surfactant concentration exceeds the CMC value, the monomers assemble and aggregate to form large amphiphilic transparent micelles. Metal cations and inorganic pollutants form a bond with the head of the ionic micelle's surface, which is oppositely charged *via* electrostatic interaction [38]. The micelles formed in solution trap the target molecules on their surface, so that membrane could retain them notwithstanding the small size of the ions. Few papers dealing with boron removal by MEUF process are present in literature. Bryjak et al. [39] studied the removal of traces amounts of

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