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# Use of reverse osmosis as a polish for the cationic surfactant after electrooxidative treatment: Acute and chronic toxicity assessment



Mariana S. Paula<sup>a,b</sup>, Rodrigo P.M. Ribeiro<sup>b</sup>, Alexandre A. Cerqueira<sup>a</sup>, Fábio Merçon<sup>b</sup>, Daniele Maia Bila<sup>c</sup>, Louise da Cruz Felix<sup>c</sup>, Mônica R.C. Marques<sup>a,\*</sup>

<sup>a</sup> Laboratório de Tecnologia Ambiental – Faculdade de Engenharia -Universidade do Estado Rio de Janeiro (UERJ), Rua São Francisco Xavier, 524, Maracanã, Rio de Janeiro, CEP 20550-900 RJ, Brazil

<sup>b</sup> Departamento de Tecnologia de Processos Bioquímicos – Faculdade de Engenharia -Universidade do Estado Rio de Janeiro (UERJ), Rua São Francisco Xavier, 524, Maracanã, Rio de Janeiro, CEP 20550-900 RJ, Brazil

<sup>c</sup> Instituto de Química, and Laboratório de Engenharia Sanitária – Faculdade de Engenharia -Universidade do Estado Rio de Janeiro (UERJ), Rua São Francisco Xavier, 524, Maracanã, Rio de Janeiro, CEP 20550-900 RJ, Brazil

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#### ABSTRACT

The main purpose of this work was to evaluate the performance of a commercial reverse osmosis (RO) membrane regarding selectivity (rejection) and productivity (permeate flux) of the treatment of quaternary ammonium compounds (QAC) after electro-Fenton (EF) treatment. Pollutants treated after the EF process should be investigated for ecotoxicity, since excess ions and high conductivity are harmful to aquatic and terrestrial biota. The use of the membrane system after EF treatment acts as final polishing since some electro-oxidative treatments leave the sample with high conductivity. In this study, RO was operated with a constant flow of 1 L min<sup>-1</sup> and feed pressures of 1 MPa, 2 MPa and 3 MPa to reject ions (sodium and iron) and to decrease the level of toxicity using representative species from different taxonomic groups: freshwater algae (*Pseudokirchneriella subcapitata*), microcrustaceans (*Daphnia similis*) and lettuce seeds (*Lactuca sativa*). Experiments carried out at different pressures showed that increased pressure caused a rise in rejection and permeate flux. At the applied pressure of 3 MPa, after 180 min, conductivity removal efficiency of 83% was obtained, 85% for sodium and 99% for iron at a flow of 13.87 L/h m<sup>2</sup>. In all bioassays, the use of the membrane was efficient to decrease the toxicity by rejecting the ions. The microcrustacean tested was the most sensitive organism, while alga was the most tolerant organism. The germination of lettuce seeds and the relative growth rate of the radicle after the combined EF + RO process was satisfactory.

### 1. Introduction

In recent decades, global production of surfactants has grown remarkably, associated with new needs of modern society and technological advances (Di Corcia, 1998). Surfactants are considered among the most undesirable pollutants found in the aquatic and terrestrial environments (Gomez et al., 2011; Olmez-Hanci et al., 2011). In aquatic environments, intense foam formation can reduce oxygen levels, resulting in deterioration of water quality, and in addition may cause toxic effects to organisms (Koparal et al., 2006; Huang et al., 2012). Cationic surfactants, such as quaternary ammonium compounds (QACs), are organic micro-pollutants that are included in the category of emerging contaminants (Clarke and Smith, 2011). This group is widely used in several areas, such as in fabric softeners, textile industry, biocides, road construction, disinfectants, emulsifiers, hair conditioners, and cosmetics, among others (Lara-Martín et al., 2010; Koner et al., 2011). These compounds are toxic at low concentrations to various organisms and should be removed from the effluent prior to discharge into surface waters (Dantas et al., 2009; Ge et al., 2010; Di Nica et al., 2017). The main toxic effect of surfactants observed in organisms is increased permeability of the cell membrane. According to Lewis (1990), this phenomenon is caused by the interaction of the surfactants with the constituent proteins of the cell walls of the microorganisms or by the cellular denaturation, possibly leading to cell lysis.

Conventional treatments, such as physical chemistry followed by biological processes, are not efficient for the complete degradation of these organic compounds (da Silva et al., 2015). Thus it is necessary to

\* Corresponding author.

E-mail address: monicamarques@uerj.br (M.R.C. Marques).

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use more efficient processes to improve the biodegradability and decrease the toxicity of QACs.

In this scenario, the advanced electrochemical oxidation processes (EAOPs) stand out. These are divided into several groups, such as anodic oxidation (AO), electro-Fenton (EF) and photoelectro-Fenton (PEF) (Moreira et al., 2017). The EF technique is subdivided into categories according to the  $H_2O_2$  source and the catalyst used in the reaction. These processes are based on the electrochemical generation of strong oxidizing agents, such as hydroxyl radicals (•OH). The production of hydroxyl radicals occurs through the reaction between hydrogen peroxide ( $H_2O_2$ ) and ferrous ions (or other catalysts) at acid pH (Wang et al., 2016). In order to accelerate the transfer of electrons during the electrochemical reaction, support electrolytes are used, the most studied being sodium sulfate ( $Na_2SO_4$ ) (Nidheesh and Gandhimathi, 2012).

The advantages of the formation of hydroxyl radicals are the high reactivity and low selectivity in the disintegration and oxidation of organic molecules in solution. Among the few disadvantages found in EAOPs are sludge formation in some cases, low pH at the end of the process (requiring adjustment before disposal of the treated effluent), and the need for final polishing to remove the catalyst and supporting electrolyte from the solution to the legal level for discharge (Moreira et al., 2017).

The literature contains some works on the degradation of surfactants using the EF technique (Lissens et al., 2003; Panizza et al., 2013, 2016), but there is a scarcity on the evaluation of the byproducts formed, since not all the processes of electrochemical oxidation attain mineralization of the surfactants. In addition, the presence of iron ions and support electrolytes after electrochemical treatments may be harmful to aquatic and terrestrial biota. One way to evaluate the byproducts is to run ecotoxicological tests using organisms of different trophic levels.

In a study by de Paula et al. (2017), the efficiency of two oxidative processes in the degradation of a cationic surfactant was compared. The authors verified that both the ozonation and the electro-Fenton processes were efficient in degrading the surfactant. However, the by-products generated after EF reaction were more toxic to lettuce seeds compared to ozonization. Some factors may have impaired seed development and physiology, such as  $Fe^{3+}$ ,  $Na^+$ ,  $Cl^-$  and  $SO_4^{-2-2}$  ions. Thus, there is a need to investigate alternatives to reduce the conductivity of the solution treated by EF.

According to Bunani et al. (2015), parameters such as salinity and conductivity directly affect the physiology of the aquatic and terrestrial biota. Excess salinity can cause imbalance in the aquatic environment and can also affect soil properties, causing crop losses.

The most commonly used technique for retaining salts, ions and others is the membrane filtration system. The use of membrane technology has been identified as one of the most robust and flexible technologies used to improve water quality, by removing salinity and pathogens (Rahardianto et al., 2007; Reverberi and Gorenflo, 2007).

Applying polishing after the electrochemical experiments is called the combined process. As an example of a combined process, there is electrocoagulation associated with membrane filtration (Romanelli et al., 2006; Urtiaga et al., 2013; de Souza et al., 2017). The use of electro-oxidative treatments before membrane filtration has been recommended because electrochemical processes can increase the salinity of the effluents, due, among other factors, to anode oxidation (Cerqueira et al., 2014).

In response to increasingly stringent water quality regulations, reverse osmosis (RO) treatment is employed in most water recovery systems for reuse (Fujioka et al., 2013). In addition, there is growing concern about integrating membrane technology with ecotoxicological studies, with different organisms of different trophic levels, to enable more efficient evaluation of the quality of the treated sample (Palma et al., 2016).

Several species of organisms have been used in ecotoxicity tests,

generating important data for better evaluation of treated effluents. Among the main groups of aquatic organisms used in laboratory tests are bacteria, microalgae, crustaceans and fish (Costa et al., 2008). The most commonly used terrestrial organisms are earthworms, lettuce and onions (da Silva et al., 2015; Demuynck et al., 2016; Cuervo Lumbaque et al., 2017). In Brazil, freshwater aquatic species are commonly used: green algae (producers) and microcrustaceans (primary consumers) of the species *Daphnia similis*, since they are the most representative in the Southern Hemisphere. For terrestrial groups there is no unanimity regarding plant species, but for invertebrate animals, worms of the *Eisenia fetida* species are most often used.

It is recommended that the toxic effect of a sample be evaluated for more than one representative aquatic species, preferably belonging to different trophic levels of the food chain (Zagatto and Gherardi-Goldstein, 1991). For bioassays with terrestrial organisms, the most recommended is the use of lettuce seeds, because they are more sensitive, simple and economical. These bioassays are called phytotoxicity tests (Charles et al., 2011).

The objective of this work was to study the influence of a reverse osmosis system (RO) on the reduction of conductivity and toxicity from the degradation of a commercial sample containing the QACs, hexadecyltrimethylammonium chloride, treated by the EF process. The efficiency of ionic rejection of iron and sodium by atomic absorption was analyzed. Acute and chronic toxicity was determined with the microalga (*Pseudokirchneriella subcapitata*), crustacean (*Daphnia similis*) and lettuce seed (*Lactuca sativa*).

## 2. Materials and methods

#### 2.1. Test solution

The test solution came from a commercial cationic surfactant, which is composed of a 28% ethanol/water solution of quaternary ammonium salt – hexadecyltrimethylammonium chloride, donated by a cosmetics company located in the state of Rio de Janeiro. The test solutions were prepared just before experiments by diluting a certain amount of commercial sample in a defined volume of distilled water. The concentration used was 0.1 mL L<sup>-1</sup> (equivalent to 22.8 mg L<sup>-1</sup>).

The sample used in the RO tests came from advanced oxidative treatment by electro-Fenton (EF), according to the following method: the EF experiments were carried out in an alternating current electro-flocculation bench unit, composed of a 1.0 L capacity cylindrical cell and iron electrode. In this experiment, 1.0 L of the test solution was oxidized at room temperature ( $25 \pm 1$  °C) at pH 3.0, adjusted with aqueous HCl solution ( $1 \mod L^{-1}$ ). The conductivity of the medium was adjusted by the addition of Na<sub>2</sub>SO<sub>4</sub> (0.03 mol L<sup>-1</sup>). After adjusting the pH and adding the electrolyte, the electrochemical reaction was started with the addition of 1.33 g L<sup>-1</sup> (equivalent to 4 mL) of P.A. H<sub>2</sub>O<sub>2</sub> (30% w/w). The experiments were carried out under the operating conditions of the reactor with current intensity of 3 A.

### 2.2. Reverse osmosis (RO)

A commercial aromatic polyamide membrane (4040-X201—TSA; Trisep Corporation<sup>®</sup>) with 0.01169 m<sup>2</sup> area of effective permeation was used. This membrane was originally compressed with ultrapure water until the water flow was constant. Previous work showed that the water permeability of the membrane is  $1.4 \text{ Lh m}^{-2}$  bar<sup>-2</sup> (de Souza et al., 2017). The schematic diagram of the experimental setup can be seen in Rodrigues Pires da Silva et al. (2016).

In this experiment, the pressure was varied at 1, 2 and 3 MPa for 3 h, with a recirculation flow of  $1 \text{ Lmin}^{-1}$ . An initial volume of about 4 L was treated in the RO system (Pam Membranes). The test solution was pumped into the membrane module through a high-pressure pump. Every 30 min, the permeate samples were collected in Falcon tubes and stored under refrigeration for further analysis.

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