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Evaluation of electrocoagulation as pre-treatment of oil emulsions, followed by reverse osmosis

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ABSTRACT

This study aimed to treat oil-in-water emulsions and obtain high quality water that is suitable for reuse. To this end, pretreatment was used with electrocoagulation (EC) to minimize fouling on the membrane and initially reduce the pollutant load, followed by reverse osmosis (RO). Two different oil-in-water emulsions and two different type of current (Alternate current, AC, and direct current, DC) were analyzed. AC or DC did not affect removal efficiency, only the consumption of the electrodes (double for DC). A residence time of 6 min was found enough to reach a constant level in terms of removal efficiencies, which were, regardless of the type of emulsion, over 99.5% in turbidity, 96% in color and 92% in chemical oxygen demand (COD). The subsequent step of RO reached 100% removal of COD and absorbance, over 99.9% of turbidity, 98.9% of total dissolved solids (TDS), 99.1% electrolytic conductivity, and 99.6% of aluminum ions, achieving the limiting flux for permeate of 20 L/h m^2 at a net pressure drop through the membrane of 2.874 MPa. Over the 2 h of the experiment, there was a small permeate flux decrease.

1. Introduction

Oily wastewaters are produced by various industries and create a major ecological problem throughout the world [1]. Produced water (PW), a type of oily water, is considered to be one of the largest waste streams in the petroleum, oil and gas industry. The amount of PW generated varies during oil production: a new field produces 5–15 vol%, while at the end of its lifetime; it reaches 75–90 vol% [2]. In 2007, nearly 21 billion barrels of PW were generated in the U.S. [3].

Recent advancements have demonstrated the potential for applying available commercial modules of membranes to produced water management in the petroleum industry [4]. Results from various researchers are found in the literature. Lin and Lan [5] were one of the first evaluating membrane processes to treat oil in water emulsions, conducted experiments to examine the efficiencies of ultrafiltration and reverse osmosis processes to reduce chemical oxygen demand (COD), total suspended solids (TSS), electrolytic conductivity and turbidity, finding over 99% improvements. Alzahrani et al. [6,7] compared NF and OI membranes and reports one advantage of RO is that the standalone application of an RO membrane can remove most of the toxic substances present in produced water. Madaeni and Eslamifard [8] used reverse osmosis to treat industrial wastewater and applied pressures of 0.5–2.2 MPa. They reported that that COD, Biochemical oxygen demand (BOD), TDS, absorbance 400 nm, turbidity, SO_4^{2-}, NH_4^+ and TSS, among others, of the wastewater were decreased and removed extensively using RO membranes. The flux of permeated stream and the recovery rate were increased with the feed pressure. The rejection of significant parameters by the membrane element was around 100%.

Because these results are so promising, reverse osmosis was chosen in this study as the treatment for the oily emulsions prepared in this study to simulate produced water. The technology could have received the raw wastewater without treatment, as shown in study by Jeshi and Neville [9], that reports that the rejection of reverse osmosis membranes is over 99% regardless of the feedstream oil concentration (valid from 0.15 to 50% of oil content). However, it is well known that RO systems can suffer from fouling of the membranes [10]. Jeshi and Neville [9] recognize that the membranes are expected to foul faster membrane at high oil contamination. It is recognized that RO would need extensive pretreatment to be feasible for the treatment of oily wastewaters [11].

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Fouling remains a major barrier to widespread implementation of membrane processes [3], and recently, Alzahrani and Mohammad [4] have concluded that currently there is no commercial or fabricated RO membranes to treat produced water without pretreatment steps.

The fouling of polymeric membranes typically forms by inorganic and organic materials present in the wastewaters, adhering to the surface and pores of the membranes and resulting in deterioration of performance (reduction of the permeation flux). In this study, to avoid the problem of excessive fouling in the RO membrane, some measures were taken. The most important of these was pretreatment of the emulsions with electrocoagulation (EC) technology, as suggested by Bensadok [12], who has concluded that electrocoagulation would be perfectly appropriate as a pretreatment step prior to a membrane process.

EC generates aluminum hydroxide in situ, which raises the pH of the treated effluent [13], and operating membrane technology at high pH values may eliminate the potential for some membrane fouling, since the solubility of some organic compounds that contribute to fouling increases with increasing pH [4]. This concept was reported in an actual application using RO membranes at high pH levels to treat produced water containing 60 mg/L of free/dispersed oil [14].

EC has a long history as a wastewater treatment method, but it has never been accepted as a mainstream method [15]. Nevertheless, EC has been used successfully to treat special wastewaters, such as nutrient enriched wastewaters, oil wastes, dye solutions, textile wastewaters, alone or in combination with another process. Zhao et al. [16] used batch electrocoagulation to treat paper mill wastewater with iron/aluminum electrodes and current densities of 100 and 150 A/m^2, and obtained COD removals between 32% and 68%, concluding that electrocoagulation can be very effective and capable of improving the paper mill wastewater quality downstream of the biological treatment.

Moussavi et al. [17] reported electrocoagulation as promising to treat petroleum-contaminated groundwater; by using iron/aluminum/steel electrodes, pH 4–11 and a current density of 20–180 A/m^2, up to 95.1% removals of total petroleum hydrocarbon were achieved in batch experiments and 93.4% in continuous ones. Valero et al. [18] used 50 A/m^2 to treat almond wastewater with iron electrodes and achieved 81% COD removal, concluding that currently electrocoagulation is already an effective technology for treating wastewater from the almond industry. Other hybrid processes involving electrocoagulation have been proposed too. For example, Mavrov et al. [19] used electrocoagulation followed by microfiltration to successfully remove heavy metals cations and anions from wastewater. In a continuous process, at 48 A/m^2, selenium reduction was 98.7%, arsenium, 99.9%, copper and lead more than 98.0% and zinc and cadmium more than 99.9%.

It is claimed that EC offers several advantages compared to standard coagulation methods including: lower chemical addition; capability to treat a wide variety of water contaminants; ability to treat finer colloidal particles due to charge inactivation; smaller sludge volumes; low power consumption; small operating costs and reduced operational footprint [10].

The aim of this work was to obtain treated effluent with a water quality high enough (i.e. very low turbidity, conductivity, COD and SS) to be suitable for use. To this end, pretreatment was used with electrocoagulation, to minimize fouling on the membrane, followed by reverse osmosis. Two different oil-in-water emulsions were used to assess whether the nature of the oil interferes in the process. In electrocoagulation, the operational parameters analyzed were: current density, type of current (DC and AC), type of electrode (aluminum and iron) and residence time. Electrode consumption was measured for all experiments. In reverse osmosis, the effect of operating pressure was analyzed (most precisely the net driving pressure through the membrane) on the efficiency of the process for removal of the remaining pollutants.

2. Experimental

All chemicals, solvents and reagents used in the course of experiments were of analytical grade with high purity. All the experimental analysis and storage procedures were made according to standard methods for The Examination of Water and Wastewater [20].

2.1. Preparation of emulsions

Water-in-oil emulsions were prepared using commercial lubricant oil (Havoline Texaco SAE 20W-50®) and petroleum (supplied by Petrobras-Brazil), in order to verify the influence of nature of oil. The experiments were conducted considering a volume (working volume) of 0.0018 m^3 of emulsion. For the water phase, distilled water was used. The oil concentration (crude oil or lubricant oil) was maintained at 1 g/L. Since ionic conductivity is crucial for electrochemical processes, sodium chloride was added at concentration to increase the electric conductivity of the mixture. Sodium chloride was selected because of the high mobility of the sodium ions and the benefit of depassivation from the chloride ions [21]. The concentration of sodium chloride was kept fixed at 1.5 g/L as suggested by author Bensadok et al. [12]. pH was not adjusted for the experiments, only measured, to analyze the performance of oil emulsions without prior interventions in its composition.

In order to stabilize the emulsions, commercial emulsifiers ALKEST 60 TW® and ALKEST SP 60® were added to the mixtures at individual concentrations of 0.1 g/L. The hydrophilic agent chose is the commercial Alkest TW 60®, which is soluble at water and has a high value in the index of hydrophilic and lipophilic balance (HLB). The lipophilic agent chosen is the commercial Alkest SP 60®, which has low HLB index, causing it to act as dispersing or solubilizing the oil present in the mixture. Emulsification was carried out by vigorous mechanical agitation of 10,000 rpm for 600 s (10 min).

2.2. Electrocoagulation system

In the study, it was used a DC power supply equipment (Tecnopetron, 0–20.0 V, 0–20.0 A) and a AC power supply (Tecnopetron, 0–20.0 V, 0–20.0 A, 60 Hz frequency). All electrochemical experiments were conducted at room temperature and pressure (25 °C and 0.1 MPa).

Experiments were run in a batch reactor consisting of a glass beaker of total volume 0.003 m^3 equipped with 4 pairs of cathode-anodes connected, all made of the same material (aluminum or iron) and installed in parallel to each other. Each electrode is 0.064 m, width × 0.1 m height × 0.002 m thickness and the distance between cathode and anode is 1 cm, which is also the distance between each adjacaent pair. The total effective electrode area is 0.0135 m^2 for each pair of cathode-anode.

In the present study, current densities used were 18.5, 37.03 and 55.5 A/m^2 for a treated volume of 1.8 L. Besides current density, the other operational parameters varied in the system were type of electrode (aluminum and iron), type of applied current (DC and AC) and residence time in the reactor (1–18 min).

The electrodes are dipped into the tank containing oil-in-water emulsions. The reactor is filled by a peristaltic pump. The power supply is turned on and the experiment proceeds. Once completed the residence time for the experiment, the power supply is turned off, and the pump pumps the effluent into the following tank, that serves as sedimentation tank.

The treated effluent is left there for 5 min in and then is collected by the bottom of the sedimentation tank. Finally, the treated