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Total petroleum hydrocarbon degradation by hybrid electrobiochemical reactor in oilfield produced water

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ABSTRACT

The crude oil drilling and extraction operations are aimed to maximize the production may be counterbalanced by the huge production of contaminated produced water (PW). PW is conventionally treated through different physical, chemical, and biological technologies. The efficiency of suggested hybrid electrobiochemical (EBC) methods for the simultaneous removal of total petroleum hydrocarbon (TPH) and sulfate from PW generated by petroleum industry is studied. Also, the factors that affect the stability of PW quality are investigated. The results indicated that the effect of biological treatment is very important to keep control of the electrochemical by-products and more TPH removal in the EBC system. The maximum TPH and sulfate removal efficiency was achieved 75% and 25.3%, respectively when the detention time was about 5.1 min and the energy consumption was 32.6 mA/cm^2. However, a slight increasing in total bacterial count was observed when the EBC compact unit worked at a flow rate of average 20 L/h. Pseudo steady state was achieved after 30 min of current application in the solution. Also, the results of the study indicate that when the current intensity was increased above optimum level, no significant results occurred due to the release of gases.

1. Introduction

In oil and gas production activities, additional water is injected into the reservoir to sustain the pressure and achieve greater recovery levels. Both formation water and injected water are produced along with hydrocarbon mixture (Abdelwahab et al., 2009). At the surface, processes are used to separate hydrocarbons from the PW. Then, PW has a mixture of soluble and insoluble constituents. Effects of PW components on the environment are as follows: (i) increase in the salinity, (ii) dispersed and soluble oil contribution in ecosystems, (iii) inclusion of other compounds from treating chemicals, (iv) higher concentration of heavy metals than in seawater, and (v) the presence of radio nuclides (Fakhru'l-Razi et al., 2009). 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% (Gomes de Lima et al., 2009).

Various techniques have been investigated for the removal of hydrocarbons from saline water, including, electrochemical oxidation (Margarida et al., 2010) biodegradation (Diaz et al., 2002; Ziagova and L.-K M, 2007), adsorption onto activated carbon (Marcilly, 2003), membrane processes (Di Bella et al., 2015), and advanced oxidation processes (Jiade et al., 2008). These techniques suffer from several technical and economic disadvantages. Although, biodegradation is an environmentally friendly process, the rate of hydrocarbon biodegradation is considerably slow and thus requires long reaction times in the range of days. Therefore, considering the extent and volume of produced water, employing complicated system to remove petroleum hydrocarbons from oilfield produced wastewater requires extremely large treatment facilities (Li et al., 2006a,b).

The greatest challenge in the area of petroleum pollutants bioremediation is to develop a process that will ensure the interaction of biocatalyst with the complex hydrocarbon by overcoming the pollutant complexity (Chandrasekhar and Venkata Mohan, 2012). Electrochemical flow cell is a variant of the conventional coagulation process in which coagulant agents are generated in situ through the dissolution of a sacrificial anode by applying current between the anode–cathode electrodes (Erick et al., 2011). Electrobiochemical (EBC) has an efficient and promising process in treating various wastewater contaminants, such as reactive dyes, azo dyes (Tyagi et al., 2014), industrial and petroleum refinery wastewater (Shammas and Wang, 2010).

Key points for the development of clean technologies are the energetic, economic and space required aspects. So, removal of soluble petroleum hydrocarbons from produced water using the electrochemical biological processes is promising to overcome the disadvantages of electrochemical and biodegradation treatment separately. Therefore, running the EBC flow cell at a laboratory scale was required to determine its optimum experimental conditions for treating real PW. Here, we evaluated the use of EBC unit to remove TPH as a novel approach for compact spaces. This paper would provide a very useful reference for the high salinity wastewater remediation and TPH degradation in aqueous solution with utilizing inert material electrodes followed by biofilter. The influence of the electrochemical zone, bacterial efficiency,

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energy consumption and TPH concentration on degradation rate was tested.

2. Materials and methods

All chemicals used in this study were analytical grade. TPH was determined calorimetrically according to immunoassay method (TPH pocket colorimeter™ II test kit, HACH Company). Deionized water (18.2 MV cm) obtained from a Veolia water system was used in all the experiments. DR 890 colorimeter and DR200 reactor from HACH Company were used for direct detections.

2.1. Reactor design

EBC flow cell with volume 1.7 L was manufactured with cooperation technicians of Civil and Environmental Engineering Department, Northeastern University, Boston, MA, USA. Two Ti-based mixed metal oxide (Ti/MMO) electrodes and biofilter (5.2 × 5.2 × 5.0 cm (W × L × T)) were used to enhance oxidation–reduction and biological degradation of TPH in flow cell (see Fig. 1). The EBC experiments have been carried out in a bench-scale plant. Ti/MMO electrodes with dimensions of 5.6 × 5.2 × 0.5 cm (W × L × T) were used as electrode materials at a spacing 3.9 cm in all experiments. Prewashed gravels with water and hydrochloric acid several times were used as biofilter media. These media were maintained in a flow of PW to construct natural selection of their biofilm (El-Masry et al., 1995; Mousa, 2012). An average flow velocity was maintained by gravity through 200 L storage tank of PW directly from crude oil production sites. Before application of current, the whole system was washed for an hour by PW. A DC linear power source with constant volt (10–30 V) during treatments was applied. All the experiments were conducted at room temperature with water temperature varying in a close range of 25–30 °C.

TPH of different water samples (P1, P2 and P3) was measured at an interval on consecutive 5 min till the pseudo-steady state was achieved that characterized by variation of less than 5% in three consecutive readings of effluent quality (Tyagi et al., 2014).

2.2. Produced water samples

PW samples were supplied by Scimitar Egypt (Issran oil field, Ras Ghareb; Egypt). PW samples analyses were carried out by total dissolved solids (TDS) that was determined experimentally according to ASTM D-1888. Conductivity and resistivity were determined according to ASTM D-1125. pH was determined experimentally according to ASTM D-1293. Salinity value was calculated upon chloride content value. Cations and anions were determined experimentally according to ASTM D-4327 using Dionex IC model DX 1100 equipped with high capacity columns. Alkaline species (CO_3, OH, and HCO_3) were determined experimentally according to ASTM D-3875. BOD_5 was determined after incubation for 5 days in tightly stoppered bottles in the dark at 20 °C and determining the oxygen consumed. Conductivities were measured at 25 °C directly in mS/cm using a digital conductivity meter (APHA, 2005). Data were expressed as means ± standard deviations (SD) of three replicates determination.

2.3. Microbiological analysis

Water samples were collected in glass bottles that have been cleansed and rinsed carefully, given a final rinse with distilled water, and sterilized by autoclave at 121 °C for 15 min. Analysis was initiated as soon as possible after collection to minimize changes in bacterial population. The enumeration of total bacterial count (TBC) in water samples (P1, P2 and P3) was done using the spreading plate technique over plate count as tryptone glucose yeast agar medium (Oxoid, Basingstoke, Hampshire, England). Additional blanks were prepared to determine contamination of plates, pipets, and room air (APHA, 2005).

It is important to be able to predict the rate at which contaminants are removed in order to design the full scale application of the technology (Rajic et al., 2016). TPH removal % was calculated by the following equation:

Removal% $= \frac{Co-Ct}{Co} * 100$ (1).where *Co* is the initial TPH concentration (mg/L) and *Ct* is TPH concentration at a defined time during treatment (mg/L). During the experiments, voltage was used as operating parameter and corresponding current was recorded and on the basis of recorded current, j was calculated. The j was calculated through the equation as follows:

$$J = \frac{current\ (A)}{Electrodesurfacearea\ (\mathrm{cm2})} \quad (2)$$

3. Results and discussion

In order to meet the needs of a regulations and sustainable development, upgrade of sewerage infrastructure is required. It is of critical importance in PW treatment designing what the composition and distribution of constituents of target water and how the degradation should be observed as efficiently as possible. Table 1 summarizes the mean values of the various parameters monitored at four selected sites of heavy petroleum production during March 2014. The data shows that the water of site 1 has the highest pollution degree with low pH but the other sites have the close degree of compositions. Further, the analytical results are illustrated in Fig. 2 depicting the variations in selected parameters at the sampling sites corresponding to total dissolved solids.

The saline wastewater in petroleum refinery is mainly from the process of oil and gas production process accompanied by various chemicals and hydrocarbons (Diaz et al., 2002). The total anions and cations concentrations were measured offline using an ion chromatography Dionex system according to the standard methods (APHA, 2005). To determine the total metal concentration, the samples were diluted 50:50 *v/v* with 4 N HNO_3 to ensure the total solubility of the metal and the results shown in Table 1. Sodium chloride shows the main compound of the PW that presents in average 56.01% that is suitable

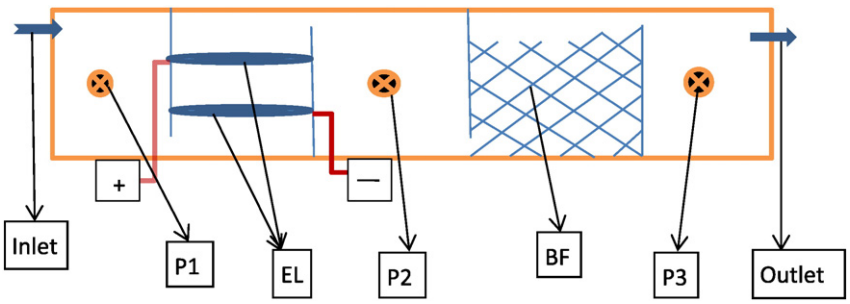

Fig. 1. Experimental setup of the EBC flow unit (two Ti/MMO electrodes (EL), biofilter (BF) and P1, P2 and P3 are sampling ports).