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Research article

Evaluation of direct and alternating current on nitrate removal using a continuous electrocoagulation process: Economical and environmental approaches through RSM

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ABSTRACT

This study aims to investigate the effects of alternating current (AC) and direct current (DC) for nitrate removal and its operating costs by using a continuous electrocoagulation (CEC) process. For this purpose, two series of 31 experiments, which were designed by response surface method (RSM), were carried out in both cases of the AC and the DC modes. In each series, the effect of selected parameters, namely, initial nitrate concentration, inlet flow rate, current density and initial pH along with their interactions on the nitrate removal efficiency as well as its operating costs, as responses, were investigated separately. According to the analysis of variance (ANOVA), there is a reasonable agreement between achieving results and the experimental data for both responses. The nitrate removal in the AC mode was slightly more efficient than that of the DC mode. In addition, the average operating costs of the DC mode, including the energy and the electrode consumption for the CEC process were achieved 54 US$/(kg nitrate removed); whereas this amount was calculated 29 US$/(kg nitrate removed) for the AC mode. Therefore, the average of the operating costs was improved more than 40% using the AC mode, which was mainly related to reduction of aluminum electrode consumption.

1. Introduction

Preserving the quality of drinking water resources presents one of the major challenges of the 21st century (Pulkka et al., 2014). Nitrate pollution, one of the serious environmental problems, has been increasing constantly in the last recent years, due to excessive application of fertilizers, discharge of municipal/industrial wastewater, animal wastes and septic systems (Azadeghan et al., 2014). Furthermore, health effects of high nitrate concentration in drinking water are most significantly linked to methemoglobinemia, also known as “blue-baby syndrome”, that affects infants (Ghafari et al., 2008). The maximum acceptable level of nitrate announced by WHO drinking water guideline is 50 mg/L as NO_3^- (WHO, 2013).

In order to keep nitrate within the approved ranges, different treatment methods in terms of physical, chemical and biological methods are used to remove this anion (Moussa et al., 2017; Pulkka et al., 2014). Applying the common physicochemical treatments such as ion exchange, electrodialysis, reverse osmosis and nanofiltration may be limited, mainly due to concentrating the pollution instead of removing it and they are also considered as expensive methods. The sensitivity of biological methods to temperature, long duration of treatment and low C:N ratios of waters are the major demerits of these approaches (Moussa et al., 2017; Yehya et al., 2015).

Electrocoagulation (EC), is reported as a promising physicochemical technology to remove various kinds of pollutants (Hakizimana et al., 2017; Moussa et al., 2017; Nariyan et al., 2018), which also represents efficient ability in nitrate removal (Emamjomeh and Sivakumar, 2009; Govindan et al., 2015; Hashim et al., 2017a; Moussa et al., 2017; Nazlabadi and Alavi Moghaddam, 2017; Pulkka et al., 2014; Xu et al., 2018; Yehya et al., 2014). Advantages of the EC process, namely, flexibility, environmental compatibility, energy efficiency, cost-effectiveness, better amenability to automation and also the ability to cope with various kinds of pollutants make it more requesting (Moussa et al., 2017; Pulkka et al., 2014). In this process, coagulant and metallic hydroxide species are generated in situ by electro-dissolution of sacrificial anode materials triggered by electric current applied through the

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electrodes (Moussa et al., 2017; Pulkka et al., 2014).

In the EC process, direct current (DC) is generally applied to produce the electric current through the electrodes. In this condition, an impermeable oxide layer may form on the cathode as well as corrosion of anodes and inhibit the effective current transport between electrodes and therefore the efficiency of the EC process may diminish. Furthermore, flows of current in one direction can reduce the lifetime of the electrodes (Ghanizadeh et al., 2016; Moussa et al., 2017). The passivity of the electrodes can be minimized using the alternating current (AC) mode in which cathode and anode can be switched periodically. Thus, the delay in cathode passivation and the anode deterioration confirms acceptable electrode life (Vasudevan and Lakshmi, 2011). The AC mode was also considered as one of the proper alternative in the EC process for different types of pollutants namely, textile dye (Tiaibaa et al., 2017), fluoride (Ghanizadeh et al., 2016), oily water (Cerqueira et al., 2014), lead and zinc (Mansoorian et al., 2014), copper (Kamaraj et al., 2013), cadmium (Vasudevan and Lakshmi, 2011) and dyes (Eyvaz et al., 2009).

Ghanizadeh et al. (2016) showed that DC mode had higher efficacy than that of AC mode for floride removal based on statistical analysis. However, Mansoorian et al. (2014) proved that the operating costs which related to energy consumption and electrode corrosion by using AC mode reduced for lead and zinc removal. Cerqueira et al. (2014) also indicated that application of AC mode of EC process for oil and grease removal promoted a lower electrode consumption as compared to using DC mode. Kamaraj et al. (2013) showed that the energy consumption reduced for copper removal as well as removal efficiency improved by using AC mode. Vasudevan and Lakshmi (2011) reported that the cadmium removal for AC mode was efficient than that of DC mode and the energy consumption reduced in AC mode, which lead to operating costs improvement. Eyvaz et al. (2009) also found that the AC mode was more efficient for dye removal.

Nitrate removal by using the EC process have been studied by other research groups (Emamjomeh and Sivakumar, 2009; Govindan et al., 2015; Hashim et al., 2017a; Kumar and Goel, 2010; Lacasa et al., 2013; Lacasa et al., 2011; Moussa et al., 2017; Nazlabadi and Alavi Moghaddam, 2017; Pulkka et al., 2014; Xu et al., 2018; Yehya et al., 2014). Also, some studies indicate that the operating cost of nitrate removal is of high expenses using the EC process (Lacasa et al., 2013; Yehya et al., 2015). However, to the best of our knowledge, evaluation of AC mode of a continuous electrocoagulation (CEC) process has not been applied for nitrate pollution to reduce its operating costs. In addition, RSM as a statistical-mathematical method could be used to evaluate the selected parameters and their interactions between responses for the CEC process (Hakizimana et al., 2017; Hendaoui et al., 2018). As is known from the literature review, RSM method is applied to evaluate EC process in batch systems for nitrate removal (Emamjomeh et al., 2017; Nazlabadi and Alavi Moghaddam, 2017), however, this method has not been simultaneously applied for nitrate removal efficiency and its operating costs using AC and DC modes in the CEC process.

The main objective of this research is an evaluation of the operating costs for nitrate removal using the CEC process. For this purpose, the electrode and the energy consumption using the AC and DC modes were compared. Hence, two series of 31 experiments were carried out in both cases through the RSM to identify the relationship between two responses (nitrate removal and its operating costs) and the selected effective parameters.

2. Materials and methods

2.1. Experimental setup

A continuous EC process was used for nitrate removal as schematically shown in Fig. 1. An electrolytic cell was made from Plexiglas with an effective volume of 2.4 L. In the cell, aluminum plate electrodes (dimension 100 × 70 × 1 mm) were connected in a monopolar parallel mode to the DC (Micro, PW4053R, 0–5A, 0–40 V) and the AC power supply (0–5A, 0–40 V, 50 Hz). The inter-electrode gaps were kept constant (10 mm) for all of the experiments. A peristaltic pump (Heidolph, PD 5201, Germany) was used to allow influent control from a reservoir tank to feed the electrolytic cell. Effluent from the electrolytic cell enters to a sedimentation tank to remove the produced suspended solids.

2.2. Experimental techniques

Sodium nitrate ($NaNO_3$) was dissolved in tap water for preparing the required initial concentration (50–250 mg/L as NO_3^-). In order to adjust the initial pH (2–10), sulphuric acid (2N) and sodium hydroxide (5N) were added to the solutions. To increase the conductivity of the solutions, sodium sulfate (Na_2SO_4) was used as a supporting electrolyte. The pH and the conductivity of the solutions were monitored during and at the end of the CEC process using a pH-meter (pH 340i, WTW, Germany) and a conductivity-meter (Cond 340, WTW, Germany). All of the experiments were conducted at the ambient temperature. At the end of each experiment, samples were taken from the effluent and were analyzed for nitrate concentration measurements. For this achievement, UV–Vis spectrophotometer (HACH, DR4000, USA) at a wavelength of 500 nm was applied according to the standard methods for examination of water and wastewater (AWWA, 1998). The performance of the CEC process was assessed in terms of the contents of nitrate concentration before and after treatment which was calculated using Eq. (1):

$$\text{Nitrate Removal Efficiency (RE\%)} = \left(\frac{C_r - C_t}{C_r}\right) \times 100 \tag{1}$$

where C_r and C_t are the nitrate concentration in raw and the treated solutions (mg/L-NO_3^-), respectively. The accuracy of the experiments was also assured using the random repetition of experiments. A scanning electron microscopy (SEM) (Seron, AIS2100, South Korea) was also used to evaluate the morphology of the electrodes after electrolysis (AC and DC modes). The SEM images of electrodes were related to those ones which were applied after all 31 runs in each current modes.

2.3. Economic analysis

Feasibility study of the EC process in large scale application, mainly depends on its cost-effectiveness. The accurate operating cost of the EC process comprises the cost of chemicals, electrode consumption, energy consumption, sludge dewatering/disposal, maintenance, pertaining to labor and fixed costs (Hakizimana et al., 2017; Hashim et al., 2017b). According to the literature, costs of electrodes and energy consumption were considerable for preliminary cost evaluation of EC process due to their major role in high expenses (Behbahani et al., 2011; Yehya et al., 2014), especially for removing low concentration of nitrate (Yehya et al., 2014). The Eq. (2) was used for estimation of these operating costs US$/(kg NO_3^- removed):

$$\text{Operating Cost (OC)} = a \times C_{energy} + b \times C_{electrodes} \tag{2}$$

where C_{energy} (kWh/(kg NO_3^- removed)) and $C_{electrodes}$ (kg Al/(kg NO_3^- removed)) are energy and electrode consumption for nitrate removal, respectively. Besides, a and b, the coefficient of the Iranian market in 2016, are described below:

Coefficient a: industrial electricity price = 0.0222 US$/kWh
Coefficient b: Wholesale Al electrode price = 1.56 US$/kg Al

The electric energy consumption of the EC process was deduced as a function of operation time through Eq. (3) as follows (Behbahani et al., 2011; Hakizimana et al., 2017; Hashim et al., 2017b):