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Research article

Energy efficient electrocoagulation using a new flow column reactor to remove nitrate from drinking water – Experimental, statistical, and economic approach

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ABSTRACT

In this investigation, a new bench-scale electrocoagulation reactor (FCER) has been applied for drinking water denitrification. FCER utilises the concepts of flow column to mix and aerate the water. The water being treated flows through the perforated aluminium disks electrodes, thereby efficiently mixing and aerating the water. As a result, FCER reduces the need for external stirring and aerating devices, which until now have been widely used in the electrocoagulation reactors. Therefore, FCER could be a promising cost-effective alternative to the traditional lab-scale EC reactors.

A comprehensive study has been commenced to investigate the performance of the new reactor. This includes the application of FCER to remove nitrate from drinking water. Estimation of the produced amount of H_2 gas and the yieldable energy from it, an estimation of its preliminary operating cost, and a SEM (scanning electron microscope) investigation of the influence of the EC process on the morphology of the surface of electrodes. Additionally, an empirical model was developed to reproduce the nitrate removal performance of the FCER.

The results obtained indicated that the FCER reduced the nitrate concentration from 100 to 15 mg/L (World Health Organization limitations for infants) after 55 min of electrolysing at initial pH of 7, GBE of 5 mm, CD of 2 mA/cm^2, and at operating cost of 0.455 US \$/m^3. Additionally, it was found that FCER emits H_2 gas enough to generate a power of 1.36 kW/m^3. Statistically, the relationship between the operating parameters and nitrate removal could be modelled with R^2 of 0.848. The obtained SEM images showed a large number dents on anode's surface due to the production of aluminium hydroxides.

1. Introduction

Nitrate is identified as one of the environmentally problematic pollutants that result from industrial and agricultural activities, as its presence at high concentration in water causes serious health problem such as the blue-baby syndrome and gastric cancer (Ghafari et al., 2008; Li et al., 2010; Vasudevan et al., 2010; Kamaraj et al., 2016). In addition, its presence in industrial waste considerably increases the volume of treated waste and a negatively influence its cohesion (Li et al., 2009). Moreover, water pollution with nitrate become a growing problem due to the wide usage of nitrogenous fertilizers, and recycling of domestic wastewater in rivers (Emamjomeh and Sivakumar, 2005; Pak, 2015).

Based on these facts and serious impacts that nitrate has on human health, the World Health Organization (WHO) has limited nitrate concentration in drinking water to 50 mg/L (for adults) (Abdallah et al., 2014; Kamaraj et al., 2016), but for infants the WHO limitations are stricter (15 mg/L) (Li et al., 2009).

Recently, to meet these limitations, many researchers have shown a great deal of interest in the electrocoagulation (EC) method as a promising alternative to remove nitrate from water due to many attractive advantages (Vasudevan and Oturan, 2014; Govindan et al., 2015; Sharma and Chopra, 2015). For instance, it does not require chemical handling, it is easy to perform, removes high concentrations of nitrate at relatively low operating cost, and

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it enables the operator to control the pollutant reduction through both the material of the electrodes and the operating parameters (Ghosh et al., 2008a; Aoudj et al., 2013). Additionally, using this technology become possible in rural areas because the required power, to perform it, could be driven from a solar panel (Chaturvedi and Dave, 2012; Kuokkanen, 2016). Therefore, the EC technology has been applied, separately or integrated with other methods, to remove nitrate from water and wastewater. For instance, Emamjomeh and Sivakumar (2005) used an EC cell, supplied with five aluminium electrodes, to remove nitrate from drinking water. The results obtained showed that the maximum nitrate removal efficiency, 90%, was achieved within 90 min of electrolysing at a current value of 2.5 A. Another study was carried out by Malakootian et al. (2011), using four pairs of aluminium electrodes, to remove nitrate from the water of Kerman province, Iran. The obtained results from this study showed that this cell was efficient enough to reduce nitrate concentration from 100 to 10.3 mg/L (89.7%) within 60 min of treatment. The combination of electrocoagulation and electro-oxidation (EC-EO) methods was applied by Naje and Abbas (2013) to remove nitrate from textile effluent. The obtained results indicated that the EC-EO method reduced the nitrate concentration by 90% within 90 min at a current value of 0.6 A. Hossini and Rezaee (2014) combined an EC cell, which supplied with two aluminium anodes and two graphite cathodes, with an air stripping system to remediate nitrate from wastewater. The obtained results demonstrated that this combined system was efficient to remove as high as 97% of nitrate within 120 min of electrolysing at a current of 0.14 A.

In spite of the acknowledged advantages of the EC method to treat a wide spectrum of pollutants from waters and wastewaters, it still has a clear deficiency in terms of both, the lack of variety in reactor design, and the availability of models for its performance (Un et al., 2013; Kuokkanen, 2016).

The current investigation therefore, has been carried out to fill a part of the gaps in the literature by using a new bench-scale electrocoagulation reactor (FCER), which utilises the concepts of flow column to mix and aerate the water, for drinking water denitrification. FCER reduces the use of external stirring and aerating devices which require extra power to work; these devices until now have been widely used in the EC reactors (especially laboratory scale ones). Therefore, FCER could be a cost-effective alternative to the traditional lab-scale EC reactors.

2. Aims and objectives

The current study has been carried out to fill a part of the mentioned gaps in literature through; firstly, application of a new EC reactor (FCER), which utilises the concepts of flow column to mix and aerate water being treated, for denitrification of drinking water. The influence of key operating parameters, such as the initial pH (from 4 to 10), current density (CD) (1, 2, and 3 mA/cm^2), the gap between electrodes (GBE) (from 3 to 10 mm), electrolysing time (t) (from 0 to 70 min), and initial concentration of nitrate (C_0) (from 50 to 150 mg/L) on nitrate removal will be investigated. Secondly, development of an empirical model to reproduce the nitrate removal performance of the FCER within the studied values of the operating parameters. Thirdly, conducting a preliminary economic study to estimate the minimum operating cost for nitrate removal using FCER. Fourthly, estimate the emitted amount of hydrogen gas from this new reactor during the denitrification of drinking water. The yieldable energy from recycling this eco-friendly gas also will be estimated. Finally, the influence of the electrolysing process on the texture of the perforated anodes will be investigated using the SEM (scanning electron microscope) technology.

3. Theory of nitrate reduction

The literature demonstrates that one of the most effective technologies for the removal of nitrate from water is the chemical denitrification with aluminium (Murphy, 1991; Emamjomeh and Sivakumar, 2005; Pak, 2015). For instance, adding of powdered aluminium reduces nitrate to nitrite to ammonia and nitrite according to the following mechanisms (Murphy, 1991; Emamjomeh and Sivakumar, 2005):

$$3NO_3^- + 2Al + 3H_2O \rightarrow 3NO_2^- + 2Al(OH)_{3(s)} \quad (1)$$

$$3NO_2^- + 6Al + 15H_2O \rightarrow 3NH_3 + 6Al(OH)_{3(S)} + 3OH^- \quad (2)$$

In The EC method, when aluminium electrodes are used, the liberated aluminium ions from the anodes reduce the nitrate to nitrogen and ammonia as follows (Koparal and Ogutveren, 2002):

$$NO_3^- + H_2O + 2e \leftrightarrow NO_2^- + 2OH^- \quad (3)$$

$$NO_3^- + 3H_2O + 5e \leftrightarrow \frac{1}{2}N_2 + 6OH^- \quad (4)$$

$$NO_3^- + 6H_2O + 8e \leftrightarrow NH_3 + 9OH^- \quad (5)$$

$$NO_2^- + 2H_2O + 3e \leftrightarrow \frac{1}{2}N_2 + 4OH^- \quad (6)$$

$$NO_2^- + 5H_2O + 6e \leftrightarrow NH_2 + 7OH^- \quad (7)$$

This complex mechanism of nitrate reduction could be summarised in the following scheme (Govindan et al., 2015) (see Scheme 1):

4. Materials and methods

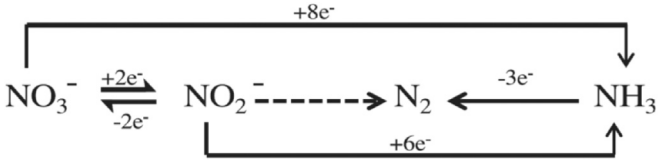
4.1. Synthetic water samples

Synthetic nitrate stock solution, 200 mg/L, was prepared by dissolving potassium nitrate (KNO_3) in deionised water. 500 mL samples with lower nitrate concentrations, ranging from 50 to 150 mg/L, were diluted from the stock solution and electrolysed at different initial such as the initial pH, CD, GBE, t, and C_0. The initial pH value of the prepared samples was adjusted to the desired value, 4 to 10, using 1 M HCl or 1 M NaOH solutions. While water conductivity was adjusted to 0.32 mS/cm using the required amount of sodium chloride (NaCl).

All chemicals were supplied by Sigma-Aldrich and used as supplied.

4.2. Batch EC reactor

In the current investigation, a new flow column reactor (FCER) has been used for water denitrification, Fig. 1. This reactor consists of a Perspex cylinder container, 25 cm in height and 10.5 cm in



Scheme 1. Reaction pathways for nitrate reduction by the EC method.