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## Effect of citric acid on the efficiency of the removal of nitrogen and phosphorus compounds during simultaneous heterotrophic-autotrophic denitrification (HAD) and electrocoagulation

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### ABSTRACT

The paper discusses the effect of electric current density and the C/N ratio on the efficiency of the removal of oxygenated nitrogen forms and total phosphorus in a bio-electrochemical sequencing batch biofilm reactor. The study was performed under anaerobic conditions, at the electric current densities of 53, 105, 158 and 201 mA m<sup>-2</sup>, the C/N ratios of 0.5, 1.0 and 1.5 and pH=7.5. For a particular electric current density, a higher efficiency of the removal of nitrogen (from 46.92( $\pm$ 1.20)% to 94.13( $\pm$ 1.50)%) and phosphorus (from 84.54( $\pm$ 1.90)% to 99.91( $\pm$ 1.60)%) compounds was achieved in the reactor fed with wastewater containing citric acid (R1) than in the control reactor without citric acid supplementation (R0). The efficiency depended on the electric current density and the dose of organic substrate. An increase in the C/N ratio of wastewater eliminated the need to apply higher electric current densities. Carbon dioxide produced during heterotrophic denitrification constituted an additional source of inorganic carbon for autotrophic bacteria. The higher total phosphorus removal efficiency in reactor R1 was caused by simultaneous electrocoagulation and higher phosphorus assimilation by the increasingly thicker biofilm.

#### 1. Introduction

High concentration of nitrogen and phosphorus in surface waters has a negative impact on water, mostly by stimulating its eutrophication (Alonso Fernández et al., 2014; Kiedrzyńska et al., 2014). Legal regulations intended to ensure a more sustainable development of the natural environment enforce action to be taken against eutrophication. The physicochemical technologies applied today to treat wastewater and sewage are not always effective or economically viable. An alternative solution is to combine biological and chemical wastewater treatment processes in a reactor with immobilized biofilm, subjected to the flow of an electric current (Kłodowska et al., 2013, 2014, 2015). Extensive research has been performed on electrocoagulation as an electrochemical process

http://dx.doi.org/10.1016/j.ecoleng.2016.06.076 0925-8574/© 2016 Elsevier B.V. All rights reserved. characterized by high efficiency of the removal of dissolved phosphorus compounds from wastewater (Chen et al., 2000; Mollah et al., 2004; İrdemez et al., 2006; Mouedhen et al., 2008; Vasudevan et al., 2009; Meas et al., 2010; Tchamango et al., 2010). In response to an electric current, electrodes in the reactor, which are most often made of aluminium or iron, undergo digestion. Metal ions thus generated bind to hydroxyl ions released on a cathode during electrolysis, thus forming metal hydroxides, which absorb soluble phosphorus compounds. The resulting agglomerates are separated from treated wastewater by sedimentation, floatation or filtration (Chen, 2004; Bayat et al., 2006; Meas et al., 2010). In contrast to conventional coagulation, electrocoagulation does not require any input of doses of coagulants because metal ions are derived from the dissolution of electrodes and their amounts are regulated by the intensity of an electric current (Koparal and Öğütveren, 2002). Moreover, the above process is distinguished by smaller quantities of generated sludge, which has superior sedimentation and dehydration qualities (Rajeshwar and Ibanez, 1997). With less energy required to run the process, it is possible to use ecological power







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**Fig. 1.** The scheme of an experimental installation with sequencing batch biofilm reactor (SBBR): (1) stainless steel cathode – disks with biomass, (2) outlet, (3) aluminium anode, (4) electric current source, (5) reactor: R0-control reactor ( $H_2$ ), R1-reactor 1 ( $H_2 + C_6 H_8 O_7$ ).

resources, e.g. fuel cells, solar panels, wind farms, etc. (Mollah et al., 2004).

The use of an immobilized biofilm reactor allows for the simultaneous removal of phosphorus compounds by electrocoagulation and nitrogen compounds by hydrogenotrophic denitrification (Krzemieniewski and Rodziewicz, 2005; Rodziewicz et al., 2011; Kłodowska et al., 2013). With the flow of an electric current, hydrogen generated on the surface of a cathode during water electrolysis becomes an internal source of energy for hydrogenotrophic bacteria, which use it to remove nitrates (Chang et al., 1999; Ghafari et al., 2009). Unlike heterotrophic denitrification, this process is highly efficient and generates negligible amounts ofsludge owing to the slow growth of biomass (Dash and Chaudhari, 2005). Furthermore, the hydrogen donor is clean, hardly soluble in water (1.6 mg L<sup>-1</sup> at 20 °C) and there is no need to remove its excess. It is a relatively inexpensive source of electrons (Lee and Rittmann, 2002; Rittmann et al., 2004; Mousavi et al., 2012).

The literature reports many studies in which researchers tried to combine the process of autotrophic denitrification with heterotrophic one (An et al., 2011; Zhao et al., 2012; Feng et al., 2013). They have demonstrated that some organic substrates may have a

positive influence on denitrification in bio-electrochemical reactors (BER). Investigations have involved such substances as methanol, glucose, starch or acetate.

No references have been found documenting research on a bioelectrochemical reactor with immobilized biofilm using citric acid as an organic substrate. Neither have the simultaneously conducted heterotrophic and autotrophic processes of denitrification and electrocoagulation been reported in the literature.

The above considerations have encouraged authors to undertake the current study, whose aim was to determine the effect of organic carbon, in the form of citric acid ( $C_6H_8O_7$ ), on the simultaneous course of heterotrophic-autotrophic denitrification and electrocoagulation.

#### 2. Materials and methods

#### 2.1. The research model

The experiment was conducted in parallel, under anaerobic conditions, in two sequencing batch bioreactors (SBBR), each with a capacity of 3.0 L and active volume of 2.0 L (Fig. 1A). Each reactor Download English Version:

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