



Research article

A 3DBER-S-EC process for simultaneous nitrogen and phosphorus removal from wastewater with low organic carbon content

Ruixia Hao ^{a,*}, Yanqing Zhou ^a, Jianbing Li ^{b,**}, Jianchao Wang ^{a,c}^a Key Laboratory of Water Quality Science and Water Environment Recovery Engineering, Beijing University of Technology, Beijing, 100124, China^b Environmental Engineering Program, University of Northern British Columbia (UNBC), Prince George, British Columbia, V2N 4Z9, Canada^c China Nuclear Power Engineering Co., Ltd., Hebei Branch, Shijiazhuang, 050000, China

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ABSTRACT

A new process was proposed by integrating a three-dimensional biofilm electrode reactor with sulfur autotrophic denitrification and electrocoagulation within the same reactor. The results indicated that under the wastewater influent condition of $\text{NO}_3\text{-N} = 30 \text{ mg/L}$, $\text{COD} = 45 \text{ mg/L}$, total phosphorus (TP) = 1.5 mg/L, hydraulic retention time (HRT) = 8 h, and $I = 400 \text{ mA}$, the $\text{NO}_3\text{-N}$ and TP removal of the proposed process reached 89.8% and 83.0%, respectively. It was observed that the electrocoagulation process improved phosphorus removal, while the simultaneous existence of heterotrophic, hydrogen, sulfur and iron autotrophic denitrifying bacteria led to enhanced and stabilized nitrogen removal. The *Sulfuritalea hydrogenivorans sk43H* and *Sulfuricella denitrificans skB26* were found as the dominant denitrifying bacteria in the electrocoagulation section and the section of biofilm electrode with sulfur filler, respectively. As compared to conventional technologies, the proposed new process can achieve simultaneous, stable and deep nitrogen and phosphorus removal from wastewater treatment plant effluent with low organic carbon content.

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1. Introduction

Due to the limitation of traditional sewage treatment process, the effluent of municipal wastewater treatment plant (WWTP) still contains a relatively high concentration of nitrate and phosphorus (Hao et al., 2013). This would restrict its direct use for water reclamation from the perspective of ecological security, and a further deep removal of nitrogen and phosphorus is of critical importance. However, WWTP effluent usually contains little organic carbon for heterotrophic denitrifying bacteria to utilize. Such problem could be addressed by biofilm-electrode reactor (BER), especially the three-dimensional BER (3D-BER) or coupling 3D-BER with other processes (Hao et al., 2013; Hussain et al., 2016). Our recent studies (Hao et al., 2016a, b) proposed an integrated process (abbreviated as 3DBER-SAD) by coupling 3D-BER with sulfur autotrophic denitrification (SAD), through the use of granular elemental sulfur (S^0) and activated carbon (AC) as the filler mixture

material in the reactor. It achieved a total nitrogen (TN) removal rate of >80% and a more stable denitrification under various operating conditions. However, this process was not effective in removing phosphorus (Nguyen et al., 2014). Electrocoagulation (EC) has been recognized as an effective and rapid phosphorus removal process with minimal sludge production (Moussa et al., 2017). It is based on the use of sacrificial electrodes (such as aluminum or iron) to produce metal ions for coagulation with nutrients that can then be removed through the combination of oxidation, flocculation and floatation (Attour et al., 2014; Chen et al., 2014; Kuokkanen et al., 2015). For example, Tian et al. (2016) examined an EC system for wastewater treatment by using a thermodynamically favorable activated carbon air cathode and a sacrificial aluminum anode, and found a phosphorus removal of up to 99% in 4 h under 1.5 cm electrode spacing and current density of 8 A m^{-2} .

In recent years, the simultaneous removal of phosphorus and nitrogen from wastewater has received increasing attention (Sun et al., 2013; Kelly and He, 2014; Wu et al., 2014; Zhang et al., 2014). However, many processes reported in previous studies were used for wastewater with relatively high organic carbon content. We recently introduced a 3D-BER filled with the mixture of sponge iron and activated carbon for simultaneous nitrogen and

* Corresponding author. College of Architecture and Civil Engineering, Beijing University of Technology, Beijing, 100124, China.

** Corresponding author.

E-mail addresses: haoruixia@bjut.edu.cn (R. Hao), Jianbing.Li@unbc.ca (J. Li).

Abbreviations

AC	activated carbon
BER	biofilm-electrode reactor
EC	electrocoagulation
HRT	hydraulic retention time
OTUs	operational taxonomic units
TN	total nitrogen
TP	total phosphorus
SAD	sulfur autotrophic denitrification
WWTP	municipal wastewater treatment plant
3DBER	three-dimensional biofilm electrode reactor
3DBER-S	3DBER integrated with sulfur autotrophic denitrification
3DBER-SAD	same as 3DBER-S, 3DBER integrated with sulfur autotrophic denitrification
3DBER-S-EC	integrated process of 3DBER-S with EC in the same reactor

phosphorus removal (Wang et al., 2014). Unfortunately, its denitrification efficiency was not improved as compared to the 3DBER-SAD process that can effectively deal with wastewater with low C/N ratio (Hao et al., 2016a). The objective of this study was then to develop an integrated process which can efficiently remove both nitrogen and phosphorus from WWTP effluent under low C/N ratio condition. We proposed to incorporate the electrocoagulation (EC) based phosphorus removal process into the 3DBER-SAD process, with the EC process being facilitated with the mixture of sponge iron and activated carbon (AC). This integrated process was abbreviated as 3DBER-S-EC. The effects of two main operating factors, including electric current and hydraulic retention time (HRT), on the performance of this new process was examined. In addition, its functional mechanism was analyzed by the understanding of its bacterial community structure using *nirS* gene-based clone library method. The results from this study would be of great importance for developing effective technology to enhance the efficiency of denitrification and phosphorus removal for WWTP effluent reclamation.

2. Materials and methods

2.1. Experimental materials

The 3DBER-S-EC experimental apparatus is a continuous up-flow reactor consisting of two reaction parts, including an EC section (with 32 cm in height) and a 3DBER-S section (with 64 cm in height) (Fig. 1). With a total effective volume of 15 L, the plexiglass reactor was cylindrical with a diameter of 25 cm. The supporting layer was 10 cm in height. The EC section was filled with a mixture of sponge iron and activated carbon (AC), while the 3DBER-S section was filled with a mixture of elemental sulfur (S^0) and AC. The volume of AC filler in each section was about 5 times as much as that of sponge iron or S^0 (Wang et al., 2014; Hao et al., 2016a), and the particle sizes of sponge iron, AC and S^0 were 5–8 mm. The reactor was connected with a DC power supply (0–60 V). The anode made of graphite rod was set in the center of the reactor, running through the EC section and ending at 10 cm below the interface of the two sections (Fig. 1). In order to increase the surface area of cathode and facilitate the attachment of microorganisms, the cathode was made of a double-layer nickle foam within polyacrylonitrile activated carbon fiber, and it was set surrounding the

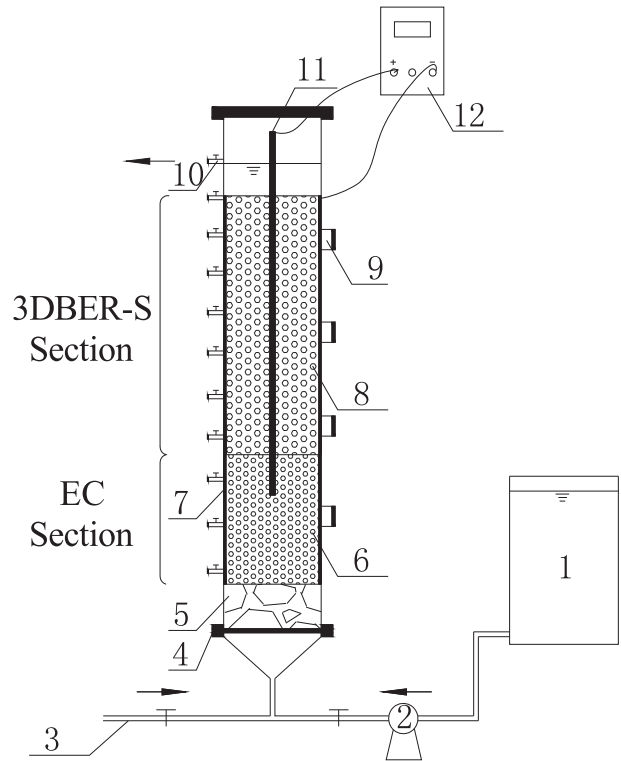


Fig. 1. Schematic diagram of 3DBER-S-EC (1-influent tank; 2-peristaltic pump; 3-washing pipe; 4-water distribution plate; 5-supporting layer; 6-filler in EC section (mixture of sponge iron and activated carbon particles); 7-cathode; 8-filler in 3DBER-S section (mixture of elemental sulfur and activated carbon particles); 9-biofilm sampling port; 10-water outlet port; 11-anode; 12-DC regulated power supply).

inner wall of the reactor through both EC and 3DBER-S sections. No short circuits were observed during the startup and operation of the 3DBER-S-EC reactor with the above configuration.

2.2. Nitrogen and phosphorus removal

2.2.1. Synthesis of simulated WWTP effluent

The influent water was a simulated WWTP effluent prepared by tap water amended with a stock solution of CH_3COONa , KNO_3 and KH_2PO_4 , while the C/N ratio was considered as the mass ratio of COD to TN. A COD, NO_3^- -N, and PO_4^{3-} -P concentration of 45, 30, and 1.5 mg/L in the influent was maintained, respectively. The C/N ratio of 1.5 is typical for WWTP effluent (Hao et al., 2013). The pH value of influent was adjusted to 7.0–7.5 using 1 mol/L HCl and 1 mol/L NaOH.

2.2.2. Operation of 3DBER-S-EC reactor

The reactor was inoculated and acclimated using anaerobic sludge taken from a WWTP in Beijing, China according to the method described in our previous studies (Hao et al., 2013, 2016a). To establish biofilm, a sludge-water with NO_3^- -N = 80 mg/L, COD = 120 mg/L, and PO_4^{3-} -P = 4 mg/L was driven by a peristaltic pump to continuously circulate through the reactor under a low flow rate (0.23 L/h), an electric current of 300 mA, and a HRT = 64 h. The biofilm was then formed on the cathode and filler surface. The microorganisms were later acclimated by gradually changing HRT to 48 h, 32 h, and 16 h, respectively. Finally, the reactor was considered to start up successfully until the NO_3^- -N removal of 3DBER-S-EC reached above 90% and the effluent was seen with no suspended sludge (Hao et al., 2013). After biofilm immobilization and acclimatization, the reactor was operated using simulated

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