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How the novel integration of electrolysis in tidal flow constructed wetlands intensifies nutrient removal and odor control

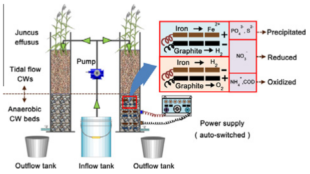
Xinxin Ju [a], Shubiao Wu [b,*], Xu Huang [c], Yansheng Zhang [a], Renjie Dong [b]

[a] *College of Water Resources & Civil Engineering, China Agricultural University, 100083 Beijing, PR China*
[b] *Key Laboratory of Clean Utilization Technology for Renewable Energy in Ministry of Agriculture, College of Engineering, China Agricultural University, 100083 Beijing, PR China*
[c] *Key Laboratory of Urban Environment and Health, Institute of Urban Environment, Chinese Academy of Sciences, 361021 Xiamen, PR China*

HIGHLIGHTS

- The coupling of tidal flow wetland with electrolysis is novelly presented.
- Electrolysis integration in wetlands has excellent effect on phosphorus removal.
- Current intensity plays an important role on nitrogen transformation.
- Odor control is achieved effectively in wetland integrated with electrolysis.
- Electrolysis has little effect on the microbial communities and abundances in CWs.

GRAPHICAL ABSTRACT



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ABSTRACT

Intensified nutrient removal and odor control in a novel electrolysis-integrated tidal flow constructed wetland were evaluated. The average removal efficiencies of COD and NH_4^+-N were above 85% and 80% in the two experimental wetlands at influent COD concentration of 300 mg/L and ammonium nitrogen concentration of 60 mg/L regardless of electrolysis integration. Effluent nitrate concentration decreased from 2.5 mg/L to 0.5 mg/L with the reduction in current intensity from 1.5 mA/cm^2 to 0.57 mA/cm^2. This result reveals the important role of current intensity in nitrogen transformation. Owing to the ferrous and ferric iron coagulant formed through the electro-dissolution of the iron anode, electrolysis integration not only exerted a positive effect on phosphorus removal but also effectively inhibited sulfide accumulation for odor control. Although electrolysis operation enhanced nutrient removal and promoted the emission of CH_4, no significant difference was observed in the microbial communities and abundance of the two experimental wetlands.

1. Introduction

The use of constructed wetlands (CWs) has become increasingly popular worldwide because of their low operating cost, good treatment efficiency, and easy maintenance (Inamori et al., 2007). However, the removal of nutrients is generally limited because of the lack of necessary oxygen to oxidize ammonium and the low sorption capacity of common substrate materials utilized for phosphorus retention (Tanner et al., 2002).

Several intensified CWs, such as artificial aerated and tidal flow CWs, have been invented to improve oxygen content in CWs (Saeed et al., 2012). Artificial aerated CWs can increase the oxygen transfer rate effectively by compressing air from the atmosphere into the wetland bed with a blower (Kadlec and Wallace, 2009). The performance of nutrient removal is intensified, and the required area is accordingly reduced. However, in consideration of energy consumption in the aeration process and the complex

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* Corresponding author. Tel.: +86 10 62737852; fax: +86 10 62737885.
E-mail address: wushubiao@gmail.com (S. Wu).

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maintenance of aerators, the large-scale application of aerated wetlands remains limited. Tidal flow constructed wetlands (TFCWs) comprise a relatively new technology that involves a novel method of oxygen transfer (Wu et al., 2011). Wastewater is rhythmically filled and drained in TFCWs and functions as a passive pump to repel and draw air from the atmosphere into the matrices (Sun et al., 2006). Consequently, the oxygen transfer rate can reach 450 g/m^2d (Wu et al., 2011). The treatment capacity of ammonium and organics is then significantly improved. However, the growth and activity of denitrifiers are often inhibited by the aerobic environment or inadequate carbon source in TFCWs; this condition stimulates nitrate in the effluent and allows for low removal of total nitrogen (TN) (Wu et al., 2011; Ju et al., 2014).

In CWs, substrates have an important function in the phosphate removal process because the adsorption and precipitation of substrates are the main removal aspects of phosphorus (Richardson, 1985; Wang et al., 2013). Many scholars have exerted efforts to study and develop efficient and practical stuffing for phosphorus removal. However, most of the substrates in CWs can only be saturated depending on time. Studies have shown that removal efficiency is good at the beginning of the operation; however, over time, the removal rate decreases significantly, and the phenomenon of phosphorus release even occurs (Brix, 1994).

Electrocoagulation has been successfully utilized to treat wastewater, particularly for phosphorus removal. With iron or aluminum as electrodes, coagulants are formed in situ through the electro-dissolution of a sacrificial anode (Cañizares et al., 2007). The metallic hydroxide flocks (ferric hydroxide) formed at the anode area can be utilized as a flocculant to deal with phosphorus in the wastewater. Simultaneously, a nondestructive chemical reaction proceeds at the cathode and produces hydrogen gas. This condition allows for the attenuation of nitrate under the condition of low carbon source; autotrophic denitrification with hydrogen as the electron donor has been studied (Lee et al., 2010; Sakakibara and Kuroda, 1993), and results have shown that the technology has a good development prospect.

NO_3^--N removal efficiency in TFCWs is not ideal because of the high oxygen content. Moreover, PO_4^{3-}-P removal in TFCWs is generally limited because of the low sorption capacity of substrate materials. Therefore, TFCWs were coupled with the technology of electrolysis in this study to fully utilize their respective advantages. A novel electrolysis-integrated tidal flow constructed wetland was developed to investigate how it intensifies nutrient removal and odor control. The dynamics of intensified nitrogen and phosphorus removal as well as the inhibitory effect of the electrolytic process on sulfide accumulation were evaluated. The contents of ammonia-oxidizing microorganisms, denitrification reductase genes, and 16SrDNA were also analyzed.

2. Methods

2.1. Laboratory-scale wetlands

The laboratory-scale wetlands utilized in this study are shown in Fig. 1. The experimental wetland integrated with electrolysis (CW II) is made of a perspex tube cylinder with internal diameter of 13 cm and length of 60 cm. This wetland was divided into two sections, with 1 L of pore water in each section. The upper section of the experimental CW is filled with zeolite (diameter of 0.5–2 mm) and operates with a tidal strategy. The lower section is filled with bio-ceramic (diameter of 3–6 mm) and integrated with electrode plates for electrolysis. The bottom section was maintained anaerobic through constant water saturation. The electrode plates (3 mm thick, 12 cm in diameter) are made of iron and/or graphite. These electrode plates are porous (diameter of 1 cm) to ensure easy water passage. The electrodes were connected by copper wires and sealed with waterproof glue. Tidal operation was generated by a peristaltic pump and an automatic drain valve controlled by a timer. Electrolysis was generated by DC-regulated power (RXN-305D, Zhaoxin, China) and controlled by a timer. A controlled wetland (CW I) without electrolysis was designed with the same configuration. These two wetland systems operate with the same tidal strategy and inflow water. Lamps (Phillips, Master SONPIA 400W, Shanghai, China) were automatically switched on as an artificial light source from 6 am to 9 pm each day. *Juncus Effusus* were planted, and the wetland was wrapped with black plastic cloth on the column sidewall to prevent periphyton formation. The system was placed in the laboratory of Bioenergy Engineering and Low Carbon Technology, China Agricultural University, at a temperature of 15 °C to 35 °C during the experiments.

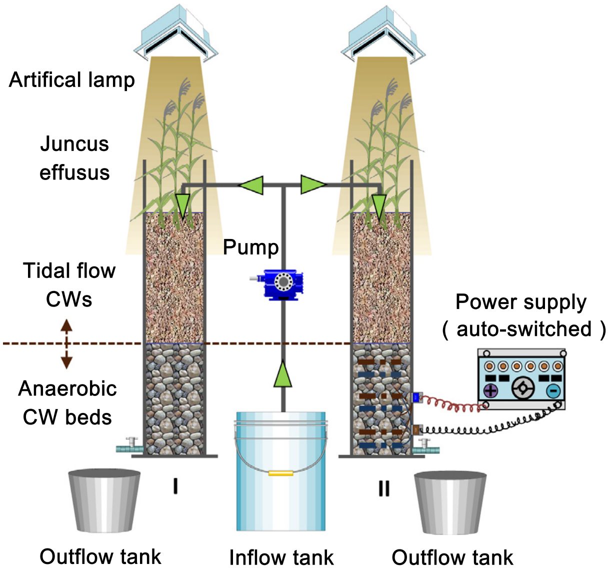

Fig. 1. Schematic of experimental laboratory-scale constructed wetlands (CWs) (System I: Non-electrolyzed contrast CW; System II: Electrolyzed experimental CW).

2.2. Experimental conditions

Synthetic wastewater containing ammonium (NH_4^+-N), chemical oxygen demand (COD) and phosphate (PO^{3-}-P) was employed to minimize variability in the experiment. Influent artificial wastewater was prepared with $C_6H_{12}O_6{\cdot}H_2O$, NH_4Cl, and KH_2PO_4 dissolved in tap water according to the required concentrations. The added compositions were COD 300 mg/L, NH_4^+-N 60 mg/L, and PO_4^{3-}-P 10 mg/L. A trace mineral solution containing EDTA-Na (0.100 g/L), $FeSO_4{\cdot}7H_2O$ (0.100 g/L), $MnCl_2{\cdot}4H_2O$ (0.100 g/L), $CoCl_2{\cdot}5H_2O$ (0.170 g/L), $CaCl_2{\cdot}6H_2O$ (0.100 g/L), $ZnCl_2$ (0.100 g/L), $CuCl_2{\cdot}5H_2O$ (0.020 g/L), $NiCl_2{\cdot}6H_2O$ (0.030 g/L), H_3BO_3 (0.010 g/L), $Na_2MoO_4{\cdot}2H_2O$ (0.010 g/L), and H_2SeO_3 (0.001 g/L) was added to the artificial wastewater (1 ml/L) in all cases (Wu et al., 2012).

The flood and drain cycle was set to occur every 8 h for the tidal operation; the flood/drain time ratio was 4 h:4 h. The condition for electrolysis followed a 4 h cycle with a voltage of 10 V. Under the initial conditions, three iron plates and two graphite plates were arranged alternately with a distance of 2 cm. The electrolytic cycle is shown in Table 1. In the nitrogen removal mode in which iron and graphite electrodes functioned as the cathode and anode, respectively, nitrogen was removed in the form of nitrogen gas using the hydrogen provided as the carbon source for denitrification. In the phosphorus removal mode in which the polarity of