Chemical Engineering Journal 237 (2014) 329–335

Contents lists available at ScienceDirect

Chemical Engineering Journal

journal homepage: www.elsevier.com/locate/cej

Achieving high-rate autotrophic nitrogen removal via Canon process in a modified single bed tidal flow constructed wetland

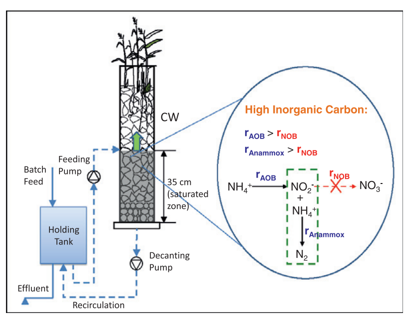
Yuansheng Hu [a,b], Xiaohong Zhao [a,c], Yaqian Zhao [a,*]

[a] UCD Dooge Centre for Water Resources Research, School of Civil, Structural and Environmental Engineering, University College Dublin, Newstead, Belfield, Dublin 4, Ireland
[b] Key Laboratory of Urban Stormwater System and Water Environment/R&D Centre for Sustainable Wastewater Treatment, Beijing University of Civil Engineering and Architecture, Ministry of Education, Beijing 100044, PR China
[c] School of Environmental Science and Engineering, Chang'an University, No. 126 of South Yanta Road, Xi'an 710054, Shanxi Province, PR China

HIGHLIGHTS

- High-rate N removal is achieved via Canon route in a modified TFCW.
- Recirculation frequency can be manipulated to control oxygen supply in the TFCW.
- Setup of a saturated zone benefits the stability of the Canon route.
- High IC provides a strong selective pressure to trigger the Canon pathway.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 3 August 2013
Received in revised form 8 October 2013
Accepted 10 October 2013
Available online 24 October 2013

Keywords:
Anammox
Constructed wetland
Inorganic carbon
Partial nitrification
Tidal flow

ABSTRACT

This study attempts to explore the Complete Autotrophic Nitrogen removal Over Nitrite (Canon) process to enhance the nitrogen removal in constructed wetlands (CWs). To achieve a stable and high-rate nitrogen removal via Canon, a single stage tidal flow constructed wetland (TFCW) system was modified by adopting internal upflow recirculation, shorter unsaturated time, and creating a pre-saturated zone. It was found that reduction of recirculation reduced oxygen supply, which favored partial nitrification and anammox process but decreased the overall nitrogen conversion rate. On the contrary, increasing the recirculation cycles enhanced the overall ammonium oxidization, but partial nitrification and anammox process could no longer be maintained. The setup of a pre-saturated zone in CW was beneficial to maintaining the stability of the Canon route. The effect of inorganic carbon (IC) on Canon was tested and the results revealed that high inorganic carbon (IC) could promote both partial nitrification and anammox activities but had no substantial effect on nitrite oxidization, which provided a strong selective method to maintain the Canon pathway. By integrating the control of oxygen supply (internal recirculation and pre-saturated zone) and high influent IC concentration, this study finally achieved total inorganic nitrogen (TIN) removal of >80% under high nitrogen loading rate of 15 g N/m^2/d from long-term stored livestock wastewater, which was characterized as having extremely low BOD_5/TN ratio of <0.2.

1. Introduction

Lower BOD_5 and higher nitrogen contents often limit classical nitrogen removal route in wastewaters. Lack of organic carbon (as a result of lower BOD_5 content) is the major factor, which hinders traditional nitrogen removal. Contradictory to the classical nitrogen removal, anaerobic ammonium oxidization (anammox) process provides an efficient and cost-effective alternative to removal nitrogen from these wastewaters without organic carbon requirement [1]. In this process, anammox bacteria convert ammonium together with nitrite (electron acceptor) directly to

* Corresponding author. Tel.: +353 1 7163215; fax: +353 1 7167399.
E-mail address: yaqian.zhao@ucd.ie (Y. Zhao).

1385-8947/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.cej.2013.10.033

dinitrogen gas in absence of oxygen [2]. It is a prerequisite for the anammox process that the antecedent nitrification process stops at nitrite by ammonia-oxidizing bacteria (AOB) (partial nitrification), i.e. the oxidation of nitrite to nitrate carried out by nitrite-oxidizing bacteria (NOB) has to be avoided [3]. This can be achieved by creating selective conditions, where AOB grow faster than NOB. Several operational parameters have been manipulated for this purpose, including temperature, dissolved oxygen (DO), free ammonia (FA) and free nitrous acid etc. Among them, low DO (~1 mg/L) and especially elevated temperature (above 25 °C) have been proven to be the strong controlling factors for partial nitrification [4].

Since distinct conditions are required for partial nitrification (aerobic) and anammox (anoxic) respectively, two-reactor (aerobic/anaerobic) system was initially adopted to facilitate these autotrophic nitrogen conversion processes [5]. Thereafter, it was found that it is feasible to integrate these two processes within a single aerobic reactor under oxygen-limiting situation, referred as Complete Autotrophic Nitrogen removal Over Nitrite (Canon) (Eq. (1)) [6,7]. It is well understood that redox stratification within the microbial aggregate (due to oxygen transfer resistance) is the major cause of the Canon process: AOB is active in the outer layer of the aggregate and producing nitrite for anammox bacteria, which exist in the inner layers and are protected from oxygen in the bulk liquid [8].

$$NH_4^+ + 0.79\ O_2 + 1.11HCO_3^- \rightarrow 0.0103\ C_5H_7O_2N(AOB) + 0.028\ CH_2O_{0.5}N_{0.15}(\text{anammox bacteria}) + 0.11NO_3^- + 0.44\ N_2 + 1.06\ CO_2 + 2.49\ H_2O \quad (1)$$

As a passively-aerated biofilm system, constructed wetlands (CWs) possess natural advantages (limited oxygen supply, redox stratification and high biomass retention, etc.) to facilitate the Canon process. Although nitrogen removal via Canon process has been reported in several studies with different types of CWs [9–13], achieving stable and high-rate autotrophic nitrogen conversion is still a challenge in such systems. One major challenge is that CWs are operated under ambient temperature, which is kinetically unfavorable to maintaining partial nitrification [4]. Also it is extremely difficult to control oxygen supply and maintain appropriate level of DO in CWs. For example, oxygen-limiting systems such as surface flow (SF, median implied oxygen transfer rate (OTR) of 1.47 g $O_2/m^2/d$) and horizontal subsurface flow (HSSF, median implied OTR of 6.3 g $O_2/m^2/d$) CWs favor both partial nitrification and anammox, but NH_4^+–N conversion is usually far from complete and overall nitrogen conversion rate is low due to the insufficient oxygen supply [12,14]. In tidal flow constructed wetlands (TFCWs), oxygen supply is greatly enhanced by the "tidal" operation (periodic saturated/unsaturated conditions) [15] and can be controlled by manipulating the duration of the saturated/unsaturated phases and the number of "tides" [16,17]. However, the oxygen supply is often substantial to trigger partial nitrification and anammox in these systems.

It was found in our previous study that oxygen supply in TFCWs could be weakened by adopting: (1) upflow mode (wastewater is introduced into the CW from the bottom and flow upward) instead of the original downflow mode (wastewater is introduced into the CW from the top and flow downward); and (2) short unsaturated time [16]. Therefore, a single bed TFCW was modified in this study to achieve the Canon route by adopting internal upflow recirculation and short unsaturated time. It was expected such adoptions would create appropriate environment for the Canon process. In addition, it was also expected that the influent inorganic carbon (IC) could be an important controlling factor for the Canon route, since IC is the carbon source for all the autotrophic microorganisms and may have significant impact on the nitrogen conversion pathway [18].

2. Materials and methods

2.1. System configuration

The modified single bed TFCW (Fig. 1a) was composed of a plexiglass column (1 m height, 9.3 cm in diameter), a holding tank (2.5 L), and two peristaltic pumps (feeding and decanting) controlled by pre-programmed timers. The first 10 cm of the bottom of the CW was filled with gravel as the support layer, followed by 60 cm of dewatered alum sludge (DAS) (particle size 1–3 cm) as the main wetland medium layer [19]. As a whole, the total depth of the CW was 70 cm and the total bed volume was 4.75 L with a working volume of 2 L (initial porosity 42%). The column was planted with common reeds.

The operation of the CW followed the "tidal flow" principal (fill-contact-drain-rest) [15]: Wastewater (from the holding tank) was rapidly (10 min) loaded to the CW in batch mode. After filling, the bed was kept saturated for a certain period of time (Table 1). Then all the wastewater was drained out rapidly (10 min) into the holding tank and the bed was allowed to "rest" (unsaturated) for a while. The whole procedure can be repeated several times a day, which creates internal recirculation of the wastewater in batch mode (Fig. 1). Finally, the treated wastewater in the holding tank was replaced with new batch of wastewater. As mentioned earlier, two modifications were adopted in this study to weaken the oxygen transfer: (1) wastewater was fed into the CW in an upflow pattern instead of the original downflow pattern (Fig. 1a); (2) a very short unsaturated period (10 min) was applied, which is usually above 1 h in the original TFCWs [15,19].

Originally, two laboratory systems (System 1 and System 2) were set up in series with the same configuration (Fig. 1a). System 1 was filled with fresh DAS, which was seeded with activated sludge from a local wastewater treatment plant. While System 2 was filled with used DAS, which was sourced from a pilot-scale TFCW system receiving long-term stored livestock wastewater characterized as low in biodegradable organic substrate [20]. After the two systems were operated with the same configuration (Fig. 1a) for 44 days, a saturated zone (1/2 of the total working volume) was adopted in System 1 (Fig. 1b): wastewater was introduced into the bed from the middle of the CW; at the end of the contact period, only half of the wastewater was drained out from the CW. As such, a stricter oxygen-limiting environment was expected to be created.

2.2. Operating strategy

There were three operating strategies tested to achieve high-rate nitrogen removal via Canon process in the modified TFCW (Table 1). Firstly, the number of the internal recirculation (N_c, times/day) was manipulated as a tool to control the oxygen supply (Phase 1). This was based on our previous finding that an initial DO of around 1 mg/L would be formed in the bed after the wastewater was fed into the CW in upflow mode [16]. Thus, the internal upflow recirculation can be regarded as an oxygenation process, and N_c acts as the times of oxygen supply. Secondly, a saturated zone was introduced in System 1 (Phase 2). This aimed at creating a stricter oxygen-limiting environment, which might be effective to maintain long-term stability of the partial nitrification and anammox activities. Thirdly, high influent IC (>200 mg/L) was applied by dosing the inflow wastewater with sodium bicarbonate ($NaHCO_3$) (Phase 3). The operational conditions in each phase were summarized in Table 1. Temperature was recorded as 18–24 °C throughout the study.