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Biological nitrogen removal in a step-feed CAST with real-time control treating municipal wastewater

Ma Juan, Peng Chengyao, Wang Li, Wang Shuying, Liu Yang, Ma Ningping, Yu Xia and Peng Yongzhen

ABSTRACT

The performance of a 18 L step-feed cyclic activated sludge technology (CAST) combined with real-time control treating real municipal wastewater was evaluated. The operation strategies employed pH and oxidation reduction potential (ORP) as on-line control parameters, which can control the durations of oxic and anoxic phases flexibly. The obtained results showed that the studied process had achieved advanced and enhanced nitrogen removal by several phases of consecutive oxic/anoxic periods. Total nitrogen in effluent was lower than 2 mg/L and the average TN removal efficiency was higher than 98%, while only requiring small amount of external carbon source. Unexpected characteristic points in pH and ORP profiles denoting the depletion of nitrate were also observed during the last anoxic phase. Denitrification rate was found to be more dependent on the system temperature compared to nitrification rate. Moreover, a stable and efficient phosphorus removal rate above 90% was achieved by using step-feed strategy which enabled the influent carbon source to be fully used and the favourable condition for phosphorus releasing to be created during the anoxic phases.

Key words | advanced nitrogen removal, cyclic activated sludge technology, real-time control, step-feed

Ma Juan
Peng Chengyao
Wang Li
Wang Shuying
Peng Yongzhen (corresponding author)
Key Laboratory of Beijing for Water Quality Science and Water Environment Recovery Engineering,
College of Environmental and Energy Engineering,
Beijing University of Technology,
100 Ping Le Yuan, Chao Yang District,
Beijing 100124,
China
E-mail: *majuan@emails.bjut.edu.cn*; *pyz@bjut.edu.cn*

Liu Yang
Ma Ningping
Yu Xia
Standard Water Ltd,
9 Story, Hongsheng Buliding, No. 98 South Road of Dongsanhuan, Chaoyang District,
Beijing 100022,
China

INTRODUCTION

Nitrogen compounds represent some of the most important pollutants in water bodies. Nitrate and ammonia in natural waters promote eutrophication which is the excess growth of aquatic algal organisms in lakes, rivers, and reservoirs. The conventional biological nitrogen removal requires a two-step process, nitrification followed by denitrification. Conventionally, nitrification is mostly achieved by the oxidation of ammonia to nitrate/nitrite, and denitrification by the subsequent reduction of nitrate/nitrite to N_2 gas under an anoxic condition at the expense of organic matter. So, the traditional biological process used in wastewater treatment to achieve nitrogen removal involves separate aerobic and anoxic phases that are generally carried out in separate bioreactors or by different aeration intervals. As more stringent effluent quality standards are imposed, advanced and cost effective techniques for nitrogen removal from wastewater become more and more important. Many modifications and novel processes have been developed and implemented for nitrogen removal from wastewater (Tchobanoglous *et al.* 2002). Cyclic activated sludge technology (CAST), a modification of sequencing batch reactor (SBR), has attracted a great deal of interest in recent years due to its performance in biological nutrient removal from wastewater and the ability of preventing sludge bulking via a selector. Being different from continuous flow activated sludge systems, various biological reactions are switchable in the same reactor. In CAST, clarifiers and flow equalization tanks are unnecessary, and thus, the capital and operational costs are much lower than those with continuous flow activated sludge systems.

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Moreover, operation conditions can be changed easily in CAST. Therefore, CAST is regarded as a cost-effective technology, especially for small wastewater treatment plants (Goronszy *et al.* 1995, 1997; Demoulin *et al.* 1997). However, conventionally operated CAST has also been limited in use by its fixed-time operating strategies since the quality of real wastewater is known to fluctuate throughout the day, which lead to unstable treatment performance. Hence, the needs for developing a novel operational scheme for CAST to enhance nitrogen removal has been increasing gradually over the last decades and further development is necessary to improve understanding of CAST operation process.

In order to achieve higher nitrification and denitrification rate, the step-feed CAST with multiple oxic (O) and anoxic (A) durations, is put forward for advanced nitrogen removal. Other than advantages seen from typical CAST, the step-feed CAST can make better use of influent COD as carbon source required in denitrification process. It suggests that, by step-feeding and multiple oxic/anoxic phases, carbon source required to denitrify nitrides formed in each oxic period is provided by the subsequent anoxic period. Moreover, step-feed strategy allows nitrification to occur under a lower organic loading in the oxic periods, which avoids the inhibition of high organic loading on autotrophic nitrifiers and saves aeration consumption to oxidize these organic matters. Along with electron transfer and alkalinity change, nitrification and denitrification are closely associated with the change of pH and oxidation reduction potential (ORP) value in the system, and some characteristic bending points will clearly appear due to the variation of rate or trend direction in pH and ORP profiles, denoting the process of biological nitrogen removal (Peng *et al.* 2002; Ingildsen & Wendelboe 2003; Akin & Ugurlu 2005; Casellas *et al.* 2006; Guo *et al.* 2007; Han *et al.* 2008). Previous investigations (Peng *et al.* 2004; Peng & Zhu 2006; Yang *et al.* 2007; Lemaire *et al.* 2008) reported that real-time control could also maintain stable partial nitrification alone. Of interest in this research is to obtain enhanced removal efficiency with real-time control for CAST system to accommodate the fluctuation of influent characteristics encountered in practice based on recognition of the specific pH or ORP bending points by the computer. In view of satisfying the Standards of Beijing Olympic Lake supplement water (TN ≤ 2 mg/L), a little dosage of external carbon source is needed.

As such, the objectives of this study were not only to investigate the advanced nitrogen removal performance in the step-feed CAST using pH and ORP as control parameters to optimize the operation, but also to study the influence of influent carbon and nitrogen ratio as well as temperature impact on the process of nitrification and denitrification. Particularly, under the condition of low influent carbon and nitrogen ratio, the trend during a typical cycle and some specific bending points in pH and ORP profiles not reported before were discussed in this research. Furthermore, the performance of phosphorus removal in the system was also investigated.

MATERIALS AND METHODS

Experimental devices

A schematic diagram of experimental equipment was depicted in Figure 1. The reactor was made of Plexiglas with a working volume of 18 L and consisted of selector and complete mix zone. The capacity ratio of selector to complete mix zone is 1: 10. An air-compressor was used for aeration. Mechanical stirrers were used to provide liquid mixing in the two zones. The feeding flow and 20% of the total returned activated sludge (RAS) were pumped to selector. In addition, pH, ORP, and dissolved oxygen (DO) sensors were installed for monitoring pH, ORP and DO in the reactor, samples were collected at intervals according to pH and ORP variations. The aeration was controlled

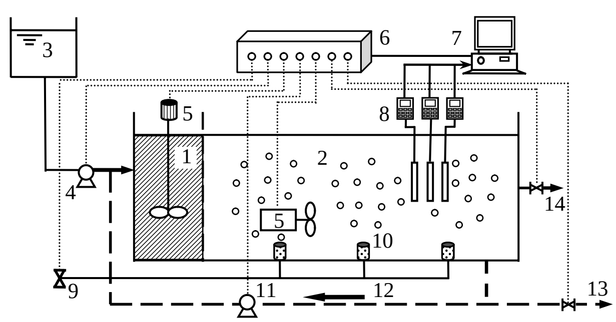

Figure 1 | Schematic diagram of experimental system and control equipment in CAST. 1. selector 2. complete mix zone 3. influent tank 4. influent pump 5. mixer 6. real-time control system 7. computer 8. DO, ORP, pH sensors 9. air pump 10. air diffuser 11. returned activated sludge pump 12. return activated sludge 13. excess sludge 14. effluent.

through manipulation of the aeration valves. RAS and feeding flow which was fixed at 144 L/d were controlled by variable speed peristaltic pumps. The mixed liquor suspended solids (MLSS) and solid retention time (SRT) were controlled at 3,000–3,500 mg/L and 10–15 days, respectively. The study was performed mostly at 23°C except tests of different temperature impact on nitrification and denitrification.

Sludge and wastewater

The inoculated activated sludge of the CAST system was taken from Jiuxianqiao wastewater treatment plant (WWTP) of OD process in Beijing city. After two-month cultivation of the activated sludge, a stable nitrogen and phosphorus removal performance had achieved and the experiment lasted two months.

The municipal wastewater used in this study was taken from the septic tank of community in Beijing University of Technology. The practical municipal wastewater used in the experiment was obtained before and after adding acetate, respectively, with low C/N ratio (COD/TN ratio: about 2.8) and high C/N ratio (COD/TN ratio: about 6.2, only used in the test of high influent C/N ratio on nitrogen removal). The C/N ratio of the raw wastewater varied between 2.1 and 4.0. The mean concentrations of chemical oxygen demand (COD), NH_4^+-N, TN and PO_4^{3-}-P in the raw wastewater were 192.3 mg/L, 69.05 mg/L, 74.5 mg/L and 5.2 mg/L, respectively. The pH value of the influent varied between 7.15 and 7.7, and the alkalinity between 200 and 420 mg/L.

Sampling and analytical methods

The routine parameters for analysis in this study included chemical oxygen demand (COD), total organic carbon (TOC), total nitrogen (TN), ammonia nitrogen (NH_4^+-N), nitrite (NO_2^--N), nitrate (NO_3^--N), phosphate (PO_4^{3-}-P), mixed liquor suspended solids (MLSS), and mixed liquor volatile suspended solids (MLVSS). Influent and effluent of the CAST were sampled every cycle for chemical analysis, and an activated sludge sample was grabbed every 2 days for concentration measurement. When the step-feed CAST operation reached a steady state, a track analysis of the entire cycle was carried out for each test in which mixed liquor samples (15 ml) were withdrawn from the CAST during the operating cycle, and at some special points, such as the end of feeding, and each non-aeration/aeration phase, and the beginning of effluent discharging. Once the samples were withdrawn from the reactor, they were immediately centrifuged at a 3,000 rpm for 2 min and filtered through 0.45 mm filter paper.

Analysis for COD, NH_4^+-N, NO_2^--N, NO_3^--N, PO_4^{3-}-P, alkalinity, MLSS and MLVSS were performed in accordance with the standard methods (APHA 1998). TOC and TN were analyzed with a Multi N/C 3400 TOC analyzer. DO, ORP and pH were measured online using DO, ORP and pH meters (WTW 340i, WTW Company), respectively.

Operation strategy

In general, a typical CAST cycle is divided into four sequences, namely fill/react, settle, draw and idle. In this case, substantial amounts of nitrite and nitrate might remain in the effluent because denitrification in selector, simultaneous nitrification and denitrification (SND) in complex mix zone and endogenous denitrification would not reduce nitrite and nitrate efficiently during react and settle phase due to shortage of electron donor. In this present work, step-feeding strategy was adopted to enhance denitrification, using the readily biodegradable organic substrates in the raw wastewater. Figure 2 shows the operational strategy. The frequencies of influent feeding included the first feeding which fulfilled in 15 min in the first aeration and following agitations except the last one. There were several oxic-anoxic combinations and at the last anoxic phase, external carbon source was added into the complex mix zone as electron donor for denitrification. Ethanol, as a potentially inexpensive, widely available electron donor for denitrification of wastewater or landfill leachate, is adopted as external carbon source in the experiment to achieve the

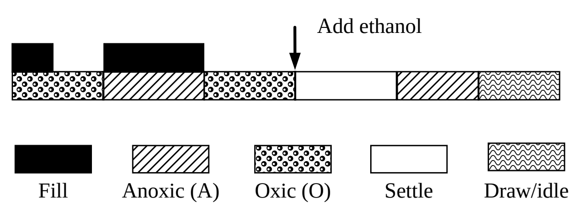

Figure 2 | Operation strategy in step-feed CAST.

advanced nitrogen removal. Aeration and agitation of the reactor ceased during the settling phase and the activated sludge was allowed to settle under quiescent conditions. During the decanting phase, the clarified supernatant was withdrawn through a valve fixed at the minimum liquid level. Both anoxic and oxic durations were not fixed but automatically programmed by real-time control, while sludge settling and effluent decanting were fixed as 60 and 10 min, respectively.

RESULTS AND DISCUSSION

The law of nitrogen removal by step-feed CAST with real time control

A typical plot of the chemical analysis in a cycle, along with pH and ORP profiles, was presented in Figure 3. The operating strategy applied to two-step feeding CAST was as follows: filling (15 min)/oxic reaction, filling /anoxic reaction, oxic reaction, ethanol addition and anoxic reaction. The observation showed that step-feed CAST

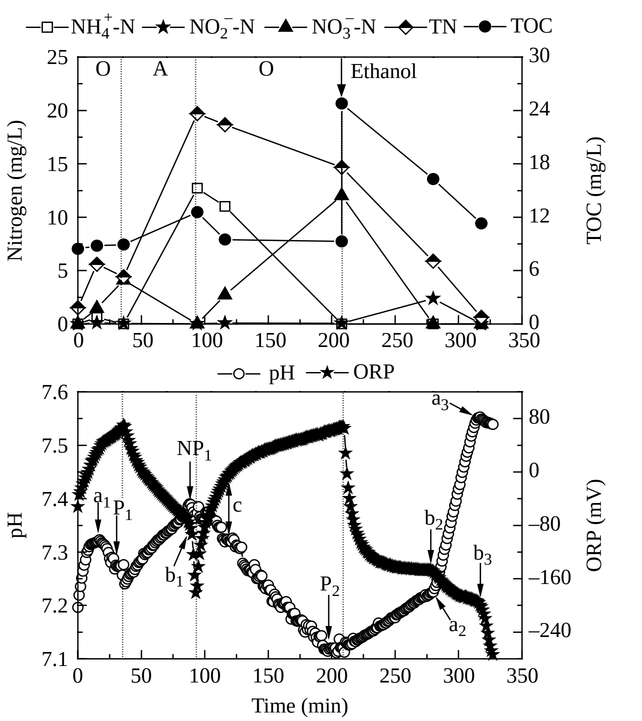

Figure 3 | Profiles of NH_4^+-N, NO_2^--N, NO_3^--N, TN, TOC and pH, ORP during a typical cycle.

combined real-time control process displayed an excellent nitrogen removal efficiency by employing several phases of consecutive oxic/anoxic periods. Total nitrogen in effluent was lower than 2 mg/L and the average TN removal efficiency was higher than 98% with influent TN (mainly in the form of ammonia) of 71.3 mg/L, while only requiring small amount of external carbon source.

In Figure 3, it was found that ORP profile varied with aeration or non-aeration regularly. In such a way, the consistent increasing or decreasing trends of ORP during the same phase could be used to diagnose operating problems resulting from aeration, which is essential for creating a favorable environment for aerobes. Compared to ORP profile, pH profile was more complex. From the beginning of the cycle to the 18th minute, a bulgy point (point a_1) in pH profile appeared. During this period, the concentration of TOC varied insignificantly and ammonia increased. Large amount of organics and small amount of ammonia were consumed by bacteria for assimilation and anabolism of new cells, which resulted in a lower ammonia concentration in practice than theoretic value. The reason why the first period defined as adsorption phase was that respiration but not anabolism dominated the system and appeared with increased ORP and pH value. The following three facts might provide an explanation why pH increased rapidly at the beginning of a cycle. First, organic acid was produced in the preceding anaerobic idle phase after complete denitrification. The organic acid in the system reduced as organic matters utilized by microbes, which resulted in an increase of pH value. Second, the anabolism and catabolism of organic matters by heterotrophic bacteria produced carbon dioxide (CO_2) and stripping of CO_2 during aerobic phase induced an increase of pH value. Third, the respiration inside bacteria consumed H^+ in the system caused pH increase. After that, nitrification dominated the system. The pH value decreased till 'ammonia valley' appeared (i.e. point P_1), which indicated the end of nitrification (Al-Ghusain & Hao 1995; Chang & Hao 1996). Accordingly, the 'nitrogen break point' (not shown for a concise graph) in ORP profile showed up. To save energy from aeration, the appearance of the pH 'ammonia valley' and ORP 'nitrogen break point' could be an effective feedback indicator for aeration adjustment to various feeding loads in the step-feed CAST.

After the completion of nitrification, influent was fed into the reactor and denitrification process began. Alkalinity produced in denitrification process resulted in pH increase, while the reduction of oxides by microbes caused a consistent decrease of ORP in the system. Since sufficient carbon source was supplied during anoxic phase, denitrification process finished at the 92th min and sulfate reduction started, thanks to the elimination of a nitrate inhibition. At this moment, the 'nitrate apex' (NP_1) (Peddie *et al.* 1990; Paul *et al.* 1998; Peng *et al.* 2003) in pH profile and the corresponding 'nitrate knee' (b_1) (Al-Ghusain & Hao 1995; Kim *et al.* 2004) in ORP profile both appeared and indicated the end of denitrification. As reported, sulfides, the products of sulfate reduction, have a great impact on ORP so that a 0.07 mg sulfide-S/l concentration increase induces a 100 mV fall of the ORP value (Plisson-Saune *et al.* 1996). As such, ORP could decline to a relatively low level in the anoxic phase due to the sulfide production.

In pH and ORP profiles of second aeration phase after the second feeding (i.e. first denitrification), point P_2 indicating the end of nitrification showed up although point c denoting complete decomposition of readily biodegradable organics. Two reasons could explain the appearance of point c. First, organics and nitrides absorbed by bacteria release to the bulk liquid resulted in a rapid pH decrease. Second, small molecular organic acid produced during the reduction of organics by microbes led to pH decline. The pH value would also decrease when the amount of CO_2 produced by anabolism exceeded the amount stripped by aeration. The main reason of ORP value keep increasing in this aerobic phase resulted from the utilization of organics by bacteria and ammonia oxidation. In Figure 3, organics degradation was observed to dominate in the reactor and nitrification hardly took place before the appearance of point c. In this period, ammonia decrease was caused by assimilation. During the time from point c to P_2, nitrification occurred in the system.

After the appearance of second 'ammonia valley' (point P_2), ethanol was added into the reactor instantly as carbon source. pH profile showed an increasing trend as nitride was denitrified. A bending point (a_3) defined as 'nitrite apex' appeared in pH profile when denitrification completed. It was also found that another inflection point a_2 presented before the appearance of 'nitrite apex' and the time point a_2 appearing was the moment when nitrate reduced to nitrite completely through detecting the concentration of chemicals. After that, denitrification of nitrite dominated the system. The pH value increased more rapidly during this phase and the slope of pH profile became steeper as a result of the higher denitrification rate of nitrite than that of complete reduction of nitrate, which enabled the appearance of point a_2. Here, point a_2 was defined as 'nitrate bow' for indicating the depletion of nitrate. Through the denitrification process, point b_2 and point b_3 on ORP profile appeared accordingly. It was a known fact that denitrification included two steps, i.e. the reduction of nitrate to nitrite and reduction of nitrite to N_2 gas. Thus, the end of denitrification should be indicated by nitrite depletion. In Figure 3, nitrite was found to accumulate during the consumption of nitrate. So, it was more reasonable to define point b_3 as 'nitrite knee' and point b_2 as 'nitrate knee'. The reason why inflection point a_2 and b_2 in pH and ORP profiles did not show up in the first anoxic duration, however, might be explained as continuous feeding and low substrate inhibited denitrification rate and the reduction rate of nitrate to nitrite lowered than that of nitrite to N_2 gas.

Effect of temperature on nitrification and denitrification in step-feed CAST with real time control

Figure 4 showed the pH and ORP profiles of advanced nitrogen removal in step-feed CAST under different temperatures. Although the three tests were performed under different temperatures, the similarity of pH and ORP response was readily apparent. Furthermore, compared to the nitrification rate, denitrification rate was found to be more dependent on the system temperature and anoxic duration was negatively affected by the temperature. Although same amount of raw wastewater was added in the first aerobic phase thus carbon source required in the subsequent anoxic phase was invariable, the effect of temperature on denitrification rate and continuous feeding style applied caused more influent fed into the reactor at lower temperature. Since the real-time control strategy was employed in the studied process to save aeration energy based on the inflection points in pH and ORP profiles,

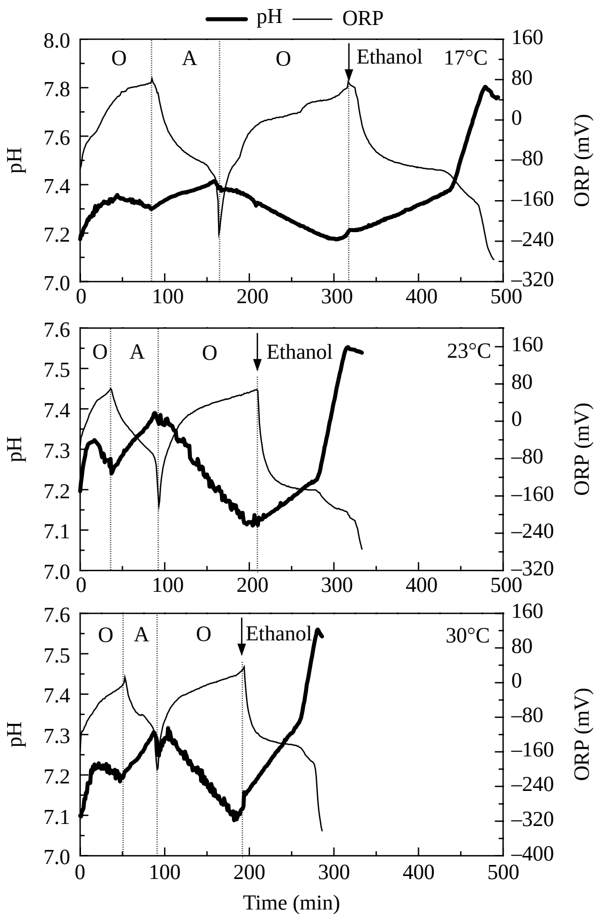

Figure 4 | Experimental pH and ORP profiles under different temperature.

feeding and aeration ceased promptly as the reaction completed. The influent added into the system exceeded the amount required at low temperature and thus the react time prolonged.

The specific nitrification/denitrification rates of the first oxic/anoxic phase and the concentration of TOC at the end of anoxic phase under different temperatures were shown in Figure 5.

It was observed from Figure 5 that the specific nitrification rate wasn't positively related to temperature. This could be explained that specific nitrification rate was influenced by lots of important factors, such as dissolved oxygen (Tchobanoglous *et al.* 2002), except for temperature. Due to the negative relationship of oxygen transfer rate and temperature, the highest specific nitrification rate was achieved under 23°C, but not under the other two temperature levels, with the same amount influent fed into

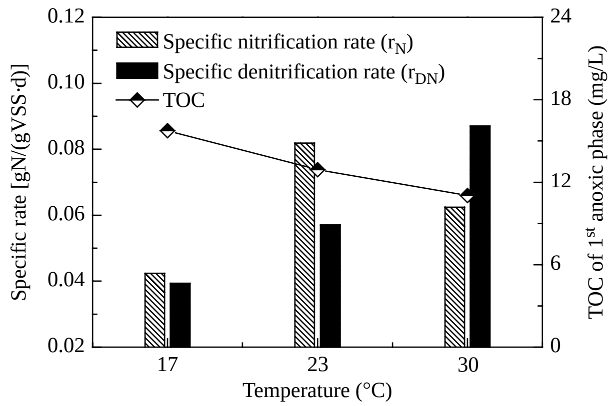

Figure 5 | Effect of temperature on both nitrification and denitrification in step-feed CAST with real time control.

the system in the first aerobic phase. On the contrary, the specific denitrification rate was improved with the increase of temperature. Because same amount carbon source was required for denitrification, the decreased specific denitrification rate and prolonged feeding time at low temperature resulted in a higher TOC concentration at the end of first anoxic phase.

Effect of high influent C/N ratio on nitrogen removal in step-feed CAST with real time control

The track analysis with high C/N ratio (about 6.2) load influent was presented in Figure 6. As shown in Figure 6, the feeding frequency increased and the duration of a cycle was shortened with influent C/N ratio increased when treating the same amount of wastewater. Compared to the treatment of low C/N ratio wastewater, ethanol dosage and the whole reaction time were saved by 80% and 20%, respectively, due to a shorter denitrification process. As a result, influent C/N ratio played an important role in improving denitrification efficiency, as higher C/N ratio enables denitrifiers to utilize more carbon in the same feeding period. It was also found that ORP profile of the first oxic phase in Figure 6 did not steadily increase as in Figure 4, though the trend was increasing. The reason was that high C/N ratio favoring phosphorus release enabled ORP value decrease significantly in selector (results not shown) and further induced fluctuation of ORP in complete mix zone at the beginning of the cycle.

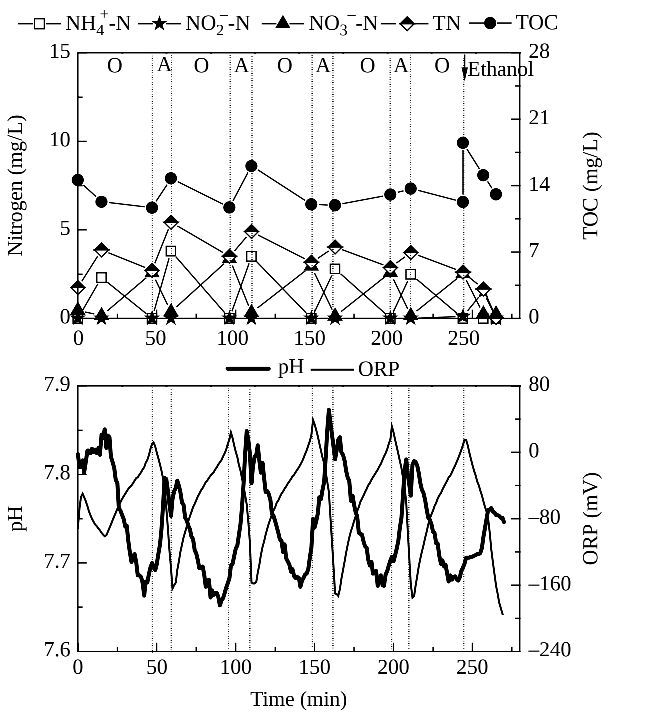

Figure 6 | Profiles of NH_4^+-N, NO_2^--N, NO_3^--N, TN, TOC and pH, ORP with high C/N influent during one typical cycle.

Performance of phosphorus removal in step-feed CAST with real time control

The phosphorus removal performance of step-feed CAST during the 30 cycles was presented in Figure 7.

Despite of the low C/P ratio of influent, a stable phosphorus removal performance was achieved with a mean removal rate above 90% by using step-feed CAST with real-time control, which resulted in approximately 0.5 mg/L phosphate in the effluent. The possible reason of efficient removal performance was that the introduction of formal anoxic mixing sequences benefited the phosphorus release in the selector. Whereas traditional operation strategy could not provide a favorable environment for phosphorus release since the complete mix zone was aerated and sludge was recycled to selector continuously during feeding time. Low carbon source made the condition even unfavorable. However, the carbon source fed into selector could be fully used with step-feed CAST process and was enough for denitrifying nitrate returned from complete mix zone and PAOs releasing phosphorus so that phosphorus release in the selector could be more efficient. Moreover, flexible distributed oxic and anoxic duration by the real-time control strategy is less convenient in causing second phosphorus release by the end of a cycle. As a result, step-feed CAST with real-time control technology exhibited good performance of not only removing nitrogen and phosphorus but also saving energy and external carbon source in wastewater treatment process.

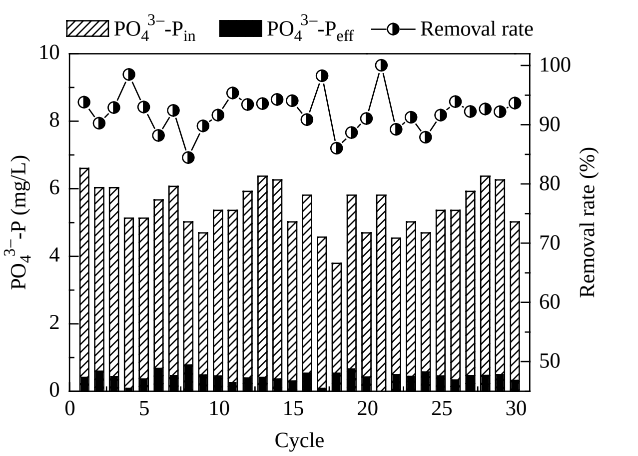

Figure 7 | Performance of phosphorus removal in step-feed CAST with real time control.

CONCLUSIONS

In a word, it could be concluded that the step-feed CAST with real-time control provided enhanced nutrient removal based on the inflection points in pH and ORP profiles to distribute the durations of oxic and anoxic phases effectively, as compared to the existing treatment performance in conventional CAST process. The system demonstrated a more flexible operation in controlling the period of the cyclic phases; the desired degree of removal could be achieved. The efficacy assessment of the bioreactor indicated that influent C/N ratio and temperature played an important role in biological nitrogen removal process. Effluent quality, however, did not vary substantially with variation in C/N ratio and temperature thanks to employing real-time control strategy in step-feed CAST, while denification rate during the nitrogen removal could be improved by increasing the C/N ratio of influent and the system.

This system could be more techno-economically viable for application in wastewater treatment for enhanced biological nutrient removal as compared to other modifications of the conventional activated sludge process, or other biological processes. Moreover, the operational concept proposed in this study is more attractive when the existing biological nutrient process is required to be retrofitted.

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