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Simultaneous Nitrogen and Phosphorus Removal from Organic Sewage Using Sequencing Batch Biofilm and Electrochemical Reactors

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The time of anaerobic, aerobic, and anoxic processes was optimized in this study as an important factor to overcome carbon source deficiency, time-consuming, high energy consumption, and high cost of sequencing-batch biofilm reactor (SBBR) processes for simultaneous removal of phosphorus and nitrogen in organic sewage treatment in the SBBR method. The anaerobic/aerobic and anaerobic/aerobic/anoxic phases of the SBBR operation for wastewater treatment were separated. In the SBBR system, K3 was used as a biofilm carrier. The results showed that the optimal time for a stable operation cycle in an SBBR system can be 5, 90, 210, 90, 20, 15 and 10 minutes for fill, anaerobic, aerobic, anoxic, settle, draw, and idle, respectively, and that the total phosphorus (TP), total nitrogen (TN), and chemical oxygen demand (COD) removal were about 97%, 94 %, and 95 %, respectively, after 90 days of operation under optimal conditions. According to the findings, under aerobic/anoxic circumstances, denitrifying phosphate accumulating organisms (DNPAOs) play a significant role in nitrate reduction and phosphorus uptake. These findings showed that in the present system, degrading bacteria, denitrifying bacteria, phosphate accumulating organisms (PAOs), and DNPAOs were enriched at the optimal time for a stable operation cycle in the SBBR system, which could primarily denitrify substances due to a lack of organic carbon. Because they can cure nitrate/nitrite and phosphate using the same carbon sources, DNPAOs can help with organic carbon competition. Finally, electrolysis based on an electrochemical method was used to post-treat residual phosphate and nitrogen ions in effluent at the end of the aerobic/anoxic period, and results showed that residual phosphate and nitrogen ions were remarkably removed in the first minutes of the electrolysis process, with contents of TP, NO2—N, and NO3—N reaching 0.78, 0.61, and 0.18 mg/l, respectively, indicating electrochemical reaction was an effective method of

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Keywords: Sequencing Batch Biofilm Reactor; Anaerobic; Aerobic, Anoxic; Nitrogen, Phosphorus, Electrochemical treatment

1. INTRODUCTION

Human waste, food, and some soap and detergents all contribute nitrogen and phosphorus to wastewater [1, 2]. Poor agricultural practices, runoff from urban areas and lawns, leaking septic systems, and sewage treatment plant outputs can all cause high phosphorus levels. The most common source of nitrate pollution is fertilizer runoff [3, 4]. According to reports, septic tank leaks and erosion of natural deposits are two other sources of nitrate contamination. Animal dung, especially cattle manure, is a major source of nitrate in wastewater, which comes in the forms of organic nitrogen, ammonia (NH_3) or ammonium (NH^{4+}), nitrate (NO^{3-}), and nitrite (NO^{3-}) (NO^{2-}) [5-7]. Total nitrogen (TN) is made up of ammonia and organic nitrogen, both of which easily sink into the groundwater via the soil and are replenished by rainwater or irrigation water [8]. Nowadays, scientific evidence shows that nitrate/nitrite exposure can cause human health effects such as increased heart rate, nausea, headaches, and abdominal cramps [9, 10]. Excess nitrates (levels >50 mg/l in drinking water) pose health hazards, including limiting oxygen transport in the bloodstream and converting hemoglobin to methemoglobin, which depletes oxygen levels in the blood, as well as being a potential cause of baby methaemoglobinaemia [11]. Phosphorus levels that are too high can harm the human body. Extra phosphorus produces changes in the body that cause calcium to be drained from the bones, making them weak [12]. Calcium deposits in the blood vessels, lungs, eyes, and heart are also harmful when phosphorus and calcium levels are high. Increased nitrate and phosphorus levels in drinking water have been related to malignancies of the colon, ovary, thyroid, kidney, and bladder, with colon cancer having the strongest link, resulting in nitrate-attributable tumors [13]. Therefore, the removal of nitrogen and phosphorus from wastewater is an essential concern because of their eutrophication effect on natural water. Chemical precipitation [14], ultra-filtration and membrane technologies [15], ion exchange [16] ionizing radiation [17], adsorption [18, 19] and SBBR systems [20, 21] are the different methods of nutrient removal which have been presented with specific advantages and disadvantages. Among them, SBBR is a widely used approach for the removal of nitrogen and phosphorus as well as COD from wastewater before it is discharged into surface or ground water, and it has gained greater attention because of advantages such as less sludge production, more flexibility, and lower cost compared to chemical methods, excellent denitrification effect, huge biomass reduction and great biofiltration activity [22, 23]. Through biological activity, ammonification converts organic nitrogen to ammonia. However, there are some disadvantages which can limit the use of the SBBR system for simultaneous removal of nitrogen and phosphorus such as a deficiency of carbon sources, time-consuming and high energy consumption and high cost of anaerobic, aerobic and anoxic processes. In addition, the high dissolved oxygen (DO) is always performed with a high nitrate residual and leads to failure in TN [24-26]. The left TN would consume carbon sources at the beginning of each cycle and cause the destruction of anaerobic conditions, resulting in weak phosphorus removal [27-29]. Therefore, it is essential to adjust the aeration conditions, especially to optimize the time of anaerobic, aerobic and anoxic processes. Therefore, this study was conducted to optimize the time of anaerobic, aerobic and anoxic processes for simultaneous removal of nitrogen and phosphorus in the SBBR system. Finally, the electrolysis based on electrochemical method was additionally applied to the post-treatment of residual phosphate and nitrogen ions in the effluent at the end of the aerobic/anoxic period.

2. EXPERIMENTAL

2.1. Fabrication and performance of SBBR system

The SBBR system used in this study consisted of a plexiglass column with a working volume of 20 l (inner diameter of 15 cm) at room temperature, which was filled with 40% volume of cylindrical K3 (AnoxKaldnes, Beijing, China) carriers as a substratum, which accelerated biofilm formation during SBBR startup [14]. The height and diameter of each K3 carrier are 12 mm and 25 mm, respectively. Per carrier, they had a specific surface area of $450m^2/m^3$. The reactor was maintained under chemostat conditions to obtain a 16-hour hydraulic retention time (HRTs) via a continuous supply of influent wastewater from the bottom of the reactor. Activated sludge was obtained from a local soybean-processing wastewater treatment plant for the inoculation procedure of a biofilm reactor in Shandong Province, China, in which starch processing ammonia wastewater was treated. The sludge retention time (SRT) was maintained at 20 days. In this study, the nitrogen loading rate was 0.2 kg NH^{4+}-N/(m^3day). The initial concentration of mixed liquor suspended solids (MLSS) in the SBBR system was 2.5 g/L.

Synthetic wastewaters were fed to the SBBR system with the main components as included (per liter): 0.03 g of $NH4^+$–N, 0.45 g of chemical oxygen demand (COD), 0.01 g TP (contained K_2HPO_4, $MgSO_4·2H_2O$ and $FeSO_4·2H_2O$), 0.04 g of $CaCl_2$ and 2 ml/l of trace element solution which contained included 50 mg/l of $NiCl_2·6H_2O$, $AlCl_3$, $MnCl_2·4H_2O$, $ZnCl_2$, $CoCl_2·6H_2O$, $Na_2MoO_4·2H_2O$, H_3BO_3 and $Na_2SeO_3·5H_2O$. To satisfy the growth requirements of nitrifying bacteria, the ratio of bicarbonate to NH^+_4-N was kept constant at a value of 7.8 mg/mg for all experiments [30]. All experiments were operated with pH control (pH meter, Fisher Scientific Accumet pH Meter 900), which varied between 7.0 and 8.5. The SBBR operation for wastewater treatment was distinguished by a series of process phases such as fill, react, settle, draw, and idle [31], and was generally divided into two steps. The first step was combining anaerobic/aerobic processes so that it is beneficial for PAOs to accumulate and start-up to make biofilm on semi-suspended bio-carriers [32]. The anaerobic/aerobic procedures were sequentially performed at a cycle, including fill, anaerobic reaction, aerobic phase, draw, and settling and idle, which were adjusted to be consistent with sedimentation of deferred activated sludge by adjusting the C/N ratio of 4:1. In the second step, anaerobic/anoxic/aerobic processes were performed to enhance the amount of the denitrifying DNPAOs, thereby caused to form the highly structured, massive, and uniform biofilm. The anaerobic/anoxic/aerobic processes were sequentially conducted on a cycle, consisting of fill, anaerobic reaction, anoxic phase, aerobic phase, draw, and settling and idle. An electromagnetic stirrer (JJ-1, JinYi, China) was used to mechanically agitate mixers to keep the biomass in suspension for mixing during the anaerobic and anoxic processes. The optimal rapid stirring speed was 100r/min.

Finally, electrolysis using an electrochemical technique was used to remove leftover phosphate and nitrogen ions in effluent as a post-treatment. A pair of Cu electrodes was used in the electrochemical cylindrical batch reactor (length of 17 cm and width of 10 cm). The electrodes were separated by 1cm. Electrolysis studies were carried out with a constant voltage of 24 V supplied by an adjustable power supply (B&K Precision, USA) and a current density of 8.6–10 A/m2. Because of its positive redox potential, slow exhaustion rate, high oxidizing power, and ease of maintenance, Cu was chosen as the electrode [18].

2.2. Characterization

On-line probes were used to measure DO levels (CPF 81, CPF 82 and OXYMAX-W COS-41, Endress-Hausserw, Weil am Rhein, Germany). DO concentrations were kept at 0.2 mg/l for anaerobic processes, 2.5 mg/l for aerobic processes, and 0.5 mg/l for anoxic processes. Finally, samples were taken and examined to determine the concentration of mixed liquid suspended solids (MLSS) and the sludge volume index (SVI) values. COD was determined using a COD analyzer (CTL-12, Chengde Huatong, China), and TN and $NH4^+$–N were measured by the fast digestion-spectrophotometric method (Nessler's Reagent Spectrophotometry, HJ/T 399-2007). The determination of the nitrite level was performed using visible spectrophotometry using N-(1-naphthyl) ethylenediamine dihydrochloride at a maximum wavelength of 540 nm. The TP levels were determined according to the Standards in Measurements and Testing (SMT) Programme extraction protocol and according to the acid molybdate spectrophotometric method. SVI and MLSS were performed in accordance with standard methods [33].

3. RESULTS AND DISCUSSION

3.1. Performance of SBBR system to treatment of NH_4^+–N, TP and COD

Figure 1 depicts the performance of the SBBR system after two months of NH4+–N, TP, and COD treatment. The elimination of NH4+–N, TP, and COD is found to be 62%, 63%, and 82%, respectively, following the first phase under MLSS of 3800 mg/l and a reduction of SVI d from 130 to 70 ml/g. Meanwhile, a thin clear yellow biofilm coating was seen on suspended carriers. After the second phase of SBBR, the removal of NH^{4+}–N, TP, and COD is achieved at 97.6%, 89%, and 91%, respectively, by decreasing settling time, increasing organic loading rate, lengthening the starvation period, and shortening the anoxic reaction and sludge discharge durations. During second phase, the MLSS was adjusted to a range from 3400 to 5000 mg/l, and SVI reduced from 45 to 53 ml/g. The density of biofilm was increased to 1.010 g/cm^3 with an average biofilm water content of 96.5% to 98.2%. Moreover, the influent water quality shows several fluctuations, indicating the great nitrification activity of the reactor for treatment of the synthetic high-strength ammonium industrial wastewater [34]. As a result, the hybrid activated sludge-biofilm system was properly created and operated, and it has high adaptability to different influent species concentrations. Furthermore, the results of the study on the operating parameters of the SBBR system indicate that the time of operation cycle for stable run of a series of process phases including fill, anaerobic, aerobic, anoxic, settle, draw, and idle is 5, 120, 210, 90, 20, 15 and 10 minutes, respectively, and a stable performance for pollutant treatment after one month for TN (97 %), TP (92 %), and COD is 5 120, 210, 90, 20, 15 and 10 minutes (97 %).

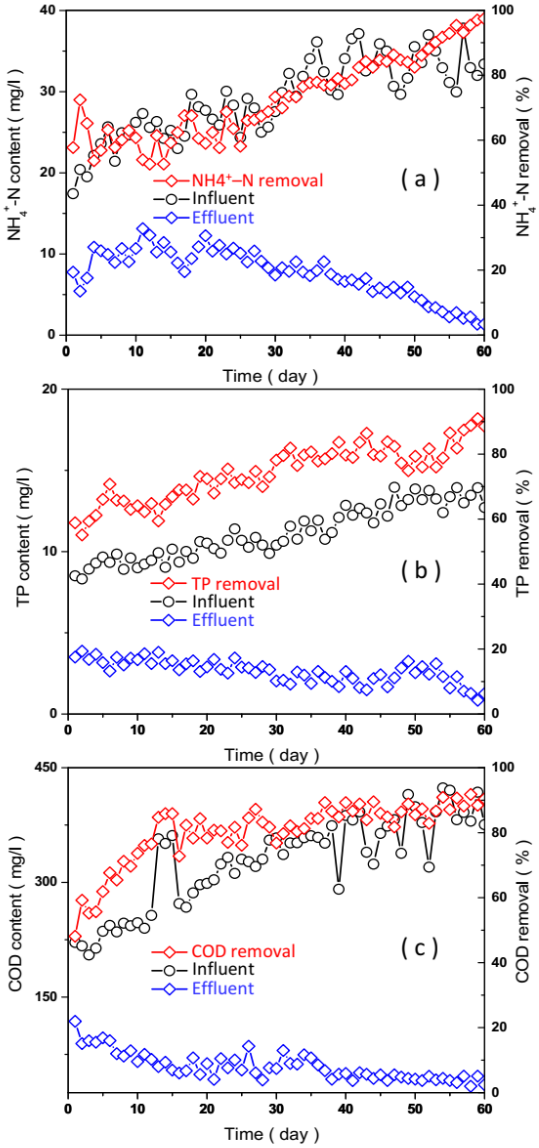

Figure 1. Performance of SBBR system after two months to treatment of (a) NH_4^+–N, (b) TP and (c) COD.

3.2. Determination of optimal time of anaerobic period

The treatment process's cost-effectiveness is heavily influenced by time and carbon source. Reduced anaerobic time can reduce the cost, energy, and time required to treat sludge, as well as consume fewer carbon sources. Meanwhile, it is considered that extending the anaerobic period can hinder the activity and growth of aerobic microorganisms [35]. In order to find the optimal time for a suitable anaerobic process, TP and COD removal were investigated in the adjusted time of the stable operation cycle as 5, 120, 180, 90, 20, 15 and 10 minutes for fill, anaerobic, draw, anoxic, settle, aerobic and idle, respectively. During the anaerobic period, the variations in TP and COD contents of wastewater specimens were analyzed every 15 minutes. The depicted results in Figure 2 reveal that the COD content is decreased quickly in the first 90 minutes then decreased slowly and then was degraded from 300 to 115.2 mg/l at the end of anaerobic phase, where organic substances including Tetrasphaera sp. and Accumulibacter sp. Predominately, they are utilized by phosphorous accumulating organisms (PAOs) to be used as cell depositor for phosphorus-uptake [36]. Normally, the more COD is consumed, the more poly-b-hydroxybutyrate (PHB) is stored [37]. During anaerobic degradation of COD, the microbial adhesion mechanism in activated sludge and microbial material system moves between the biofilm layers to create a microbial wastewater treatment membrane [38]. K_3 as a biofilm carrier may have great potential to remove pollutants from wastewater. In addition, the TP content is increased quickly in the first 90 minutes, and then gradually increased due to PAOs decomposed of intracellular polyphosphate, and unconfined energy used to produce PHB for continued metabolic activities [39, 40]. Thus, the phosphate-release is practically done with a TP content of about 46.2 mg/l after 90 minutes of anaerobic treatment, and this period demonstrates good ability for phosphate uptake and release. Therefore, 90 minutes was ascertained as the optimal period for the anaerobic process.

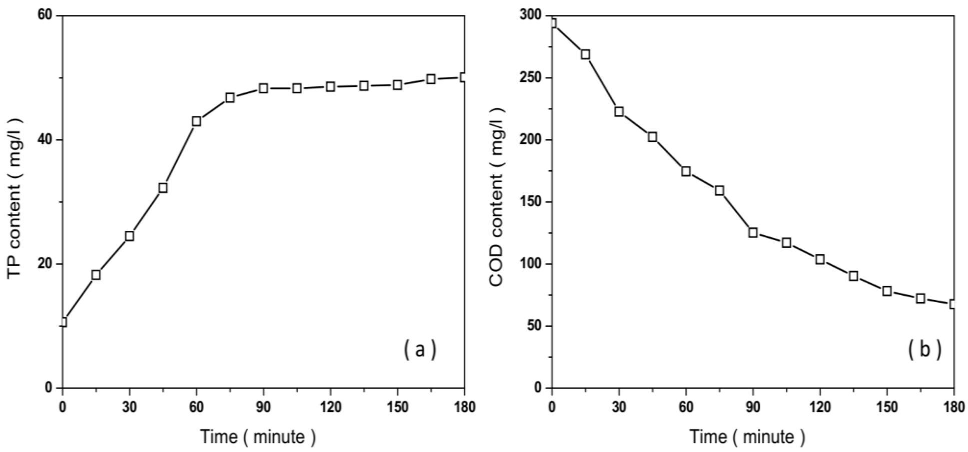

Figure 2. Variations the (a) TP and (b) COD contents of wastewater samples at anaerobic period.

3.3. Determination of optimal time of aerobic period

Optimization the aerobic period is another major parameter in the performance of the SBBR system because the adequate aeration could satisfy the conditions for efficient nitrification and phosphorus treatments [41]. For the optimal time of the aerobic process, $NH4^+$–N, TP and COD removal were studied in the adjusted time of the stable operation run as 5, 90, 300, 90, 20, 15 and 10 minutes for fill, anaerobic, draw, anoxic, settle, aerobic and idle, respectively. During the aerobic period, the variations the $NH4^+$–N, TP and COD contents of wastewater samples were characterized every 30 minutes. Figure 3 shows that NH^{4+}–N is oxidized to NO_2—N and NO_3—N by nitrosifiers and nitrifying bacteria [42], and the $NH4^+$–N content is reduced from 11.3 to 0.3 mg/l over 150 minutes..

The content of TP in wastewater is decreased from 48 to 10.2mg/L during 150 minutes. The aeration zone could continuously produce nitrite and nitrate to supply sufficient electron acceptor for DNPAOs to uptake phosphate under aerobic-anoxic conditions [36, 43]. Phosphate uptake might most probably be limited by the activity of bacteria of PAOs and DNPAOs groups and become insignificant after 180 minutes of aeration [44]. Furthermore, Figure 3 shows COD content reaches 10.2mg/l after 210 minutes and approximately exhausted. Without manure organics, the denitrification rate of phosphate uptake under anoxic condition can be more effective than under aerobic conditions. Therefore, the optimal period for aerobic phase could be kept as 210 minutes.

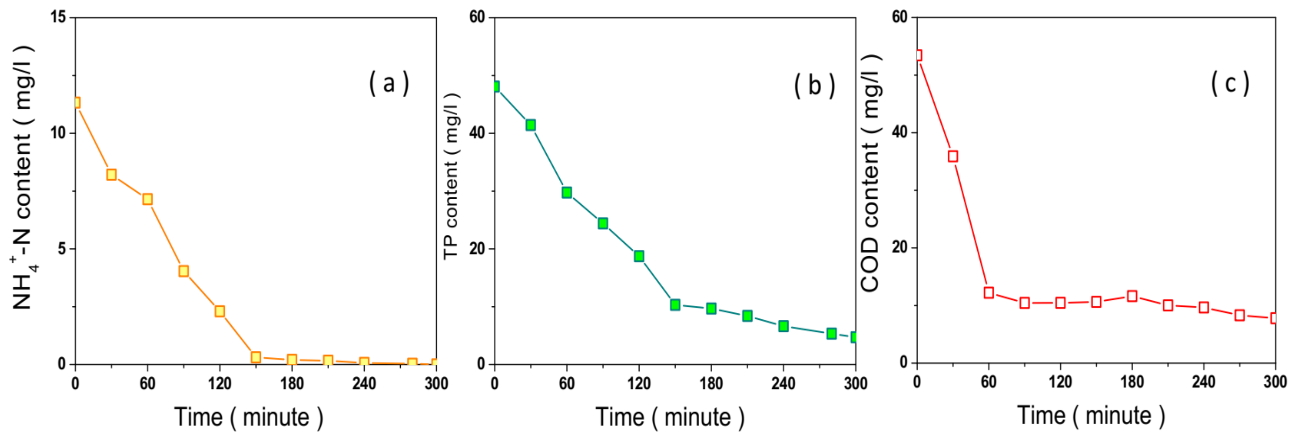

Figure 3. Variations the (a) NH_4^+–N, (b) TP and (c) COD contents of wastewater samples at aerobic period.

3.4. Determination of optimal time of anoxic period

NH^{4+}–N, TP, and COD removal were studied in the adjusted time of stable operation run of 5, 90, 210, 150, 20, 15, and 10 minutes for fill, anaerobic, aerobic, anoxic, settle, draw, and idle, respectively. During the anoxic period, the variations the NO_x^-–N and TP contents of wastewater samples were analyzed after every 30 minutes. The findings in Figure 4a and 4b show that the NO_x^-–N is reduced by bacteria in biofilm, indicating a TN content is ≥0.3 mg/L after 90 minutes. As observed, the fast treatment of TP happens in the first 90 minutes due to the fast rate of phosphate uptake in this time interval [45]. After 90 minutes, the TP content is decreased negligibly due to the low concentration of NO_3^-–N [46, 47]. Thus, 90 minutes was determined as the optimal period for the anoxic period.

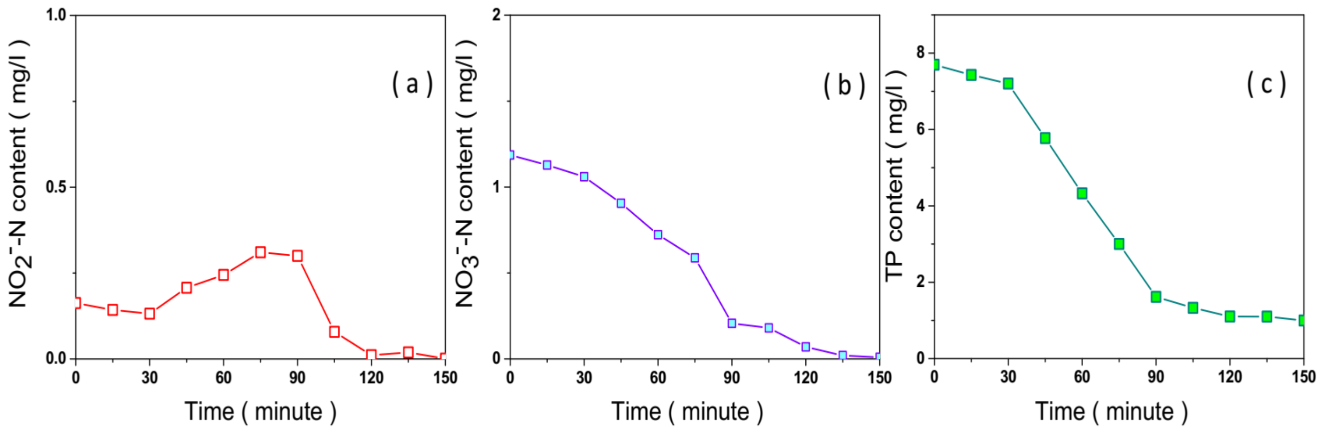

Figure 4. Variations the (a) NO_2^-–N, (b) NO_3^-–N and (c) TP contents of wastewater samples at anoxic period.

These findings indicate that the optimal time for a stable operation cycle in the SBBR system can be 5, 90, 210, 90, 20, 15 and 10 minutes for fill, anaerobic, draw, anoxic, settle, aerobic and idle, respectively.

Figure 5 shows the results of SBBR system operation under optimal conditions for 90 days, the TP, TN and COD removal is about 97%, 94% and 95%, respectively. The effluent TP, TN and COD concentration can mostly meet the Class A standard stipulated by the Chinese 'Discharge Standard of Pollutants for Municipal Wastewater Treatment Plants (GB 18918-2002) [48]. Most of the important microbial groups involved in nitrification, denitrification, biological phosphorus-removal are ammonia oxidation bacteria (AOBs), nitrite oxidation bacteria (NOBs), PAOs, DNPAOs, and other denitrifies [49]. Studies have revealed that there are large quantities of AOBs in inner layer of the biofilm, and NOBs and PAOs content in the inner layer of the biofilm are restrained due to low DO concentration all the time, where DO level and its diffusion depth could allow to growth of NOBs and PAOs within the biofilm [50, 51]. Generally, NOBs need a longer solids retention time than AOBs [49]. As a consequence, PAOs and NOBs cannot be accumulated. Meanwhile, the NOBs and PAOs contents in outer layer of the biofilm are notably enhanced because in the aerobic phase, the DO concentration is appropriate for both NOBs and PAOs [51]. Further, the transformation between aerobic and anaerobic conditions is helpful in enriching PAOs accumulating. Thus, the final phosphorus removal in the SBBR system is discharging the suspended biomass and the falling-off biofilm from the system as a surplus sludge [51]. In order to maintain stable and great phosphorus-removal, sludge containing stored polyphosphate needs to be wasted from the reactor and new cells grown to take up phosphorus from the bulk solution [49].

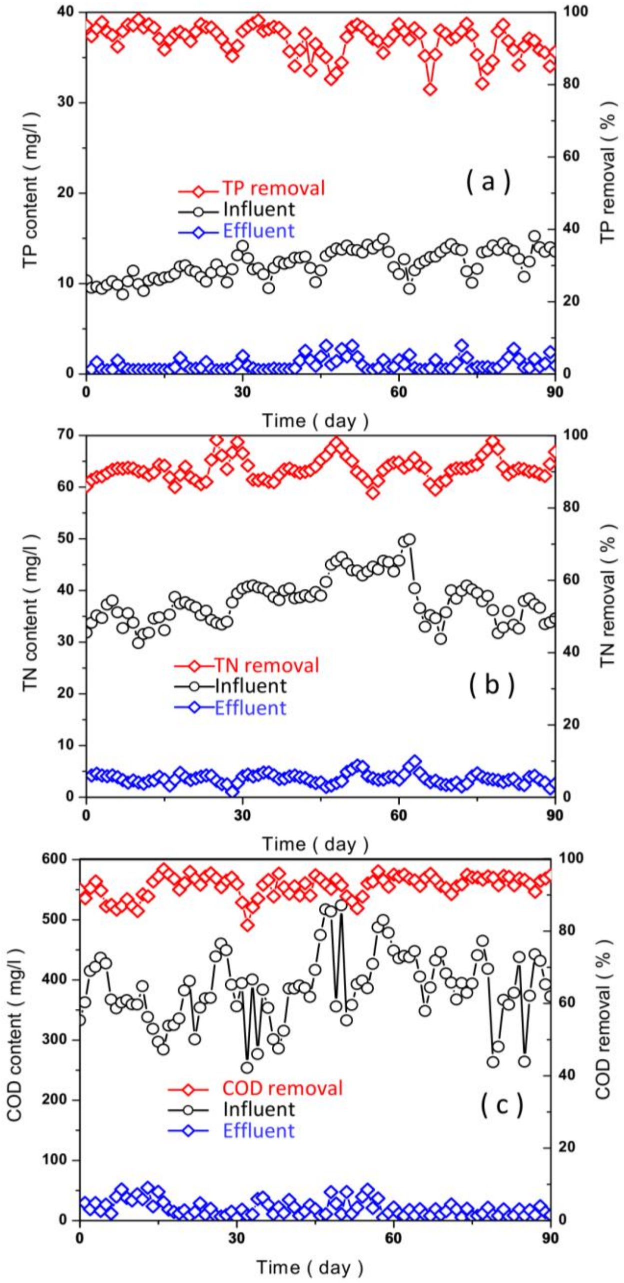

Figure 5. Results of SBBR system operation under the optimal conditions for 90 days to (a) TP, (b) TN and (c) COD removal

3.5. Characterizing mechanisms of simultaneous removal of nitrogen and phosphorus during wastewater treatment

Figure 6 shows a typical operation cycle of simultaneous removal of nitrogen and phosphorus during wastewater treatment. As found in anaerobic time, the content of COD rapidly decreased from 400 to 100.2 mg/l, and content of TP rapidly increased from 13.6 to 47.8 mg/l. The content of PAOs and DNPAOs can rapidly assimilate the phosphate coupled by the COD [43]. As depicted, the COD removal gets 75%, and the majority of the COD content would lead to more intracellular PHB, which can be stored as internal carbon source in the preceding anaerobic phase [52]. In the aerobic phase, the content of NH_4^+–N and TP exhibit a descending trend, and the content of NO_2^-–N and NO_3^-–N displays an upward trend. Nitrifiers in the aerobic phase can continuously produce nitrite and nitrate to supply sufficient electron acceptor for DNPAOs to uptake phosphate [43]. Also, in the presence of sufficient DO in the anoxic phase, the PAOs can serve as electron acceptors to uptake phosphorus, and the $NH4^+$–N converted to nitrite and nitrate [42]. However, the content of COD gradually decreased from 100.2 to 26.1 mg/l, thereby DNPAOs could mainly denitrificate substances by reason of deficiency organic carbon. The DNPAOs can assist in the competition for organic carbon because they can treat nitrate/nitrite and phosphate using the same carbon sources. Thus, during the anaerobic phase, PAOs and DNPAOs take up external carbon substrates and store them as PHB in the system, and during the anoxic phase, they can utilize nitrite or nitrate instead of oxygen as an electron accepter to remove phosphorus [53]. As a result, nitrogen and phosphorus can be simultaneously treated by integration of nitrification and denitrifying phosphorus removal. As seen at the end of the aerobic/anoxic period, NH_4^+–N is entirely oxidized to NO_2^-–N (1.9 mg/l) and NO_3^-–N (0.65 mg/l). The content of TP reaches 1.6 mg/l.

3.6. Electrochemical post-treatment of residual phosphate and nitrogen ions in final effluent of SBBR system

By considering the content of TP, NO_2^-–N and NO_3^-–N at the end of the aerobic/anoxic period, the electrochemical reaction was applied as a useful technique for guaranteeing the excellent treatment of residual phosphate and nitrogen ions from wastewater [54]. As shown in Figure 6, the residual phosphate and nitrogen ions are remarkably removed in the first minutes of the electrolysis process and after 60 minutes, the contents of TP, NO_2^-–N and NO_3^-–N reach to 0.78, 0.61 and 0.18 mg/l, respectively. This treatment is attributed to electro-coagulation mechanism which involves the generation of coagulants in situ by dissolving electrically Cu ions from the anode electrode [55, 56], and the Cu ions can perform removal of phosphorus as a follow-up reaction at the cathode [57]:

$$3\,Cu^{2+} + 2\,PO_4^{\,3-} \rightarrow Cu_3(PO_4)_2 \qquad (1)$$

For TN removal from final effluent of SBBR system by electrochemical processes as suggested mechanism [58]:

$$NO_3^- + 3\,H_2O + 5e \rightarrow \frac{1}{2}N_2 + 6OH^- \qquad (2)$$

$$NO_2^- + 2\,H_2O + 3e \rightarrow \frac{1}{2}N_2 + 4OH^- \qquad (3)$$

$$2NO_2^- + 5\,H_2O + 6e \rightarrow 3NH_3 + 7OH^- \qquad (4)$$

Therefore, it is believed that the electrochemical post-treatment of final effluent of the SBBR system using a copper electrode can be a good additional process to remove (≥99%) of TP and TN ions.

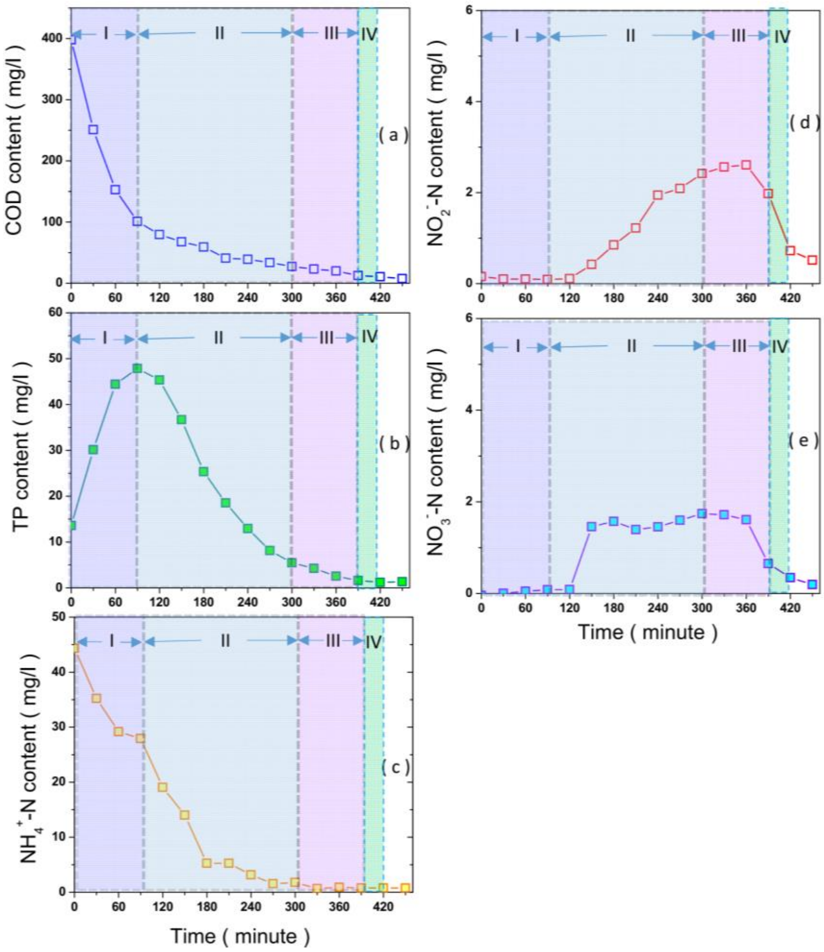

Figure 6. Typical operation cycle of removal of (a) COD, (b) TP, (c) NH_4^+–N, (d) NO_2^-–N and (e) NO_3^-–N; and (I), (II), (III) and (IV) referred to the anaerobic, aerobic, anoxic phase, and electrochemical post-treatment respectively.

4. CONCLUSION

This study was focused on finding the relatively short anaerobic/aerobic/anoxic time as the optimal and favorable time for the simultaneous removal of nitrogen and phosphorus in organic sewage treatment in the SBBR system. The results showed that the optimal time for stable operation cycle in SBBR system can be 5, 90, 210, 90, 20, 15 and 10 minutes for fill, anaerobic, aerobic, anoxic, settle, draw and idle, respectively, and results of SBBR system operation under the optimal conditions for 90 days indicated that the TP, TN and COD removal were obtained about 97%, 94% and 95%, respectively. Results showed that DNPAOs played a major role in nitrate reduction and phosphorus uptake under optimal conditions of aerobic/anoxic phases, and PAOs and DNPAOs were enriched in the present system which could mainly denitrificate substances by reason of a deficiency in organic carbon. The DNPAOs can assist in the competition for organic carbon because they can treat nitrate/nitrite and phosphate using the same carbon sources. Finally, the electrolysis based on the electrochemical method was additionally applied to the post-treatment of residual phosphate and nitrogen ions in effluent the end of the aerobic/anoxic period, and results showed that after 60 minutes, the contents of TP, NO_2^-–N and NO_3^-–N reached to 0.78, 0.61 and 0.18 mg/l, respectively, indicating electrochemical reaction was a useful technique for guaranteeing the excellent treatment of residual phosphate and nitrogen ions (≥ 99%) from wastewater.

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