A soil infiltration system incorporated with sulfur-utilizing autotrophic denitrification (SISSAD) for domestic wastewater treatment

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HIGHLIGHTS

• A SISSAD for domestic wastewater treatment is successfully developed.
• CO2 from heterotrophic aerobic respiration in AES could be used for autotrophic bacteria in ANS.
• COD and NH4+-N are removed in AES, NO3-N is reduced in ANS, TP is removed by adsorption.

1. Introduction

In China, the total amount of discharged wastewater in 2012 is 68.48 billion tons, in which domestic wastewater accounts for 67.6% (SEPA of China, 2012). It is of increasing concern for both researchers and government officials to prevent pollution of aquatic environments on domestic wastewater treatment (Wu et al., 2011). Owing to the excellent performance, the conventional activated sludge process has become the most widely used alternative for the treatment of domestic wastewater. However, the large land occupation and the high energy consumption make a wastewater treatment plant barely built in small towns or rural areas, and even more difficulties will be produced on handling wastewater in rural areas than that in cities (Ying et al., 2011). Fortunately, constructed wetland (CW), benefiting from its low cost, energy saving, simple maintenance, and higher effluent quality, has been widely applied to domestic wastewater in some rural areas (Brunch et al., 2011). However, as a natural system, the performance of CW always relies on seasonal changes and vegetations (Kouki et al., 2009).

On the other hand, subsurface wastewater infiltration treatment (SWI) is somehow superior to CW because of their higher removal efficiency and better controllability as well as space occupation (Li et al., 2013b). However, this method need to overcome the hurdle that using natural soil as a filter media will eventually cause clogging (Pozo-Morales et al., 2013). The multi-soil-layering system (MSL), filling the void with some large particle filter material such as zeolite and sand around the soil to create a filter layer, was invented to maintain relatively high removal efficiency and avoid clogging (Watasuki et al., 1993). Compared with traditional soil infiltration methods, MSL remarkably lengthened the service life and performed the removal efficiencies of 75% of TN, 80% of TP and 95% of BOD for treating restaurant wastewater (Luanmanee et al., 2001). However, the nitrate removal...
efficiency was relatively low in the MSL, mainly due to the inhibition on denitrification by aeration and lack of electron donor caused by low C/N (Masunaga et al., 2007). It has been reported that artificial aeration could improve nitrogen removal efficiency (Butterworth et al., 2013). However, an excessive aeration will consequently bring about the decreasing of both denitrification efficiency and overall nitrogen removal efficiency.

Thereby, the majority of previous soil treatment researches mainly focused on individually promoting and optimizing the heterotrophic denitrification process by artificial aeration controlling or organic carbon source addition such as wood chips and saw dust (Liu et al., 2013). In comparison with heterotrophic denitrification, anoxic autotrophic denitrification using sulfur as an electron donor exhibited its higher nitrate removal and less cell yield (Sierra-Alvarez et al., 2007), and some studies related to sulfur/limestone autotrophic denitrification (SLAD) system have been reported (Ergas et al., 2010; Read-daily et al., 2011). The sulfur/limestone autotrophic process corresponded to the principle described as following equation:

\[
55S + 20CO_2 + 50NO_3^- + 38H_2O + 4NH_4^+ \rightarrow 4C_3H_4O_2N + 25N_2 + 55SO_4^{2-} + 64H^+ + \text{Thiobacillus denitrificans}
\]

Although there are some attempts of combining aerobic stage with anaerobic one to establish a two-stage type (Chinialia et al., 2012), few researches related to incorporating sulfur-based denitrification with soil-based nitrification had been carried out.

In this study, a lab-scale SISSAD reactor was developed by integrating aerobic stage with anaerobic stage, which allows secondary effluent directly flow into sulfur-based denitrification stage without pumping, consequently reducing space occupation and energy cost. To evaluate the performance of SISSAD, the removals of COD, NH\(_4\)\(^+\)-N, NO\(_2\)\(^-\)-N, and TP in both aerobic and anaerobic stage were measured respectively. The denitrification efficiency was almost 100% in both aerobic and anaerobic stage were measured respectively.

In the anaerobic stage (ANS), the filter materials (Table 1) and mixture of sulfur granules (grain diameter 0.5–1 mm, CR, Guangfu Research Institute, China) and crushed oyster shells (S/C = 3:1 (v/v)) were filled. There were more quartz sands with larger size and volcanic filled in AES, aiming to create a relatively higher porosity in AES than in ANS. The oyster shells were obtained from local restaurant and crushed into particles (diameter 2–4 mm) by a small high-speed pulverizer (2N-04, KINGSLH Co., China) after washing and drying. AES and ANS were separated by another spongy porous membrane (30 cm × 20 cm × 1 cm) to ensure the distribution uniformity for the effluent from AES.

A glass funnel (diameter 90 mm) covered with gauze (1 mm thickness) was planted between the two stages to exactly collect the effluent from AES. The height of AES was 4 cm longer than that of ANS in order to ensure the growth of nitrifying bacteria.

2.3. Experimental procedure

2.3.1. Acclimation for nitrifying and denitrifying bacteria

For acclimation of nitrifying bacteria, an activated sludge collected from secondary sedimentation tank (Qinghe Wastewater Plant, Beijing) was cultured in a 1 L beaker fed with 5 g C\(_6\)H\(_{12}\)O\(_6\), 2 g NH\(_4\)Cl and 0.5 g KH\(_2\)PO\(_4\) (AR, Sinopharm Chemical Reagent Co., China), and aerated with air pump for 6 h every day, while another activated sludge from anaerobic tank (Qinghe Wastewater Plant, Beijing, China) for acclimation of denitrifying bacteria was cultivated in a sealed 500 mL conical flask containing 200 mL medium, which was prepared by dissolving 1 g Na\(_2\)S\(_2\)O\(_3\), 0.2 g NaHCO\(_3\), 0.4 g KH\(_2\)PO\(_4\), 0.1 g NH\(_4\)Cl, 0.1 g MgCl\(_2\)6H\(_2\)O and 0.002 g FeSO\(_4\)7H\(_2\)O (AR, Sinopharm Chemical Reagent Co., China) in distilled 200 mL water, and then incubated in a DDHZ-300 Shaker (Peiying, Taicang Experiment Equipment Co., China) at 30 °C. The medium was renewed every day to provide a proper condition for the anaerobic denitrifying bacteria growth. After 2 weeks aqueous culturing, the sediment in the conical flask was transferred into another 250 mL conical flasks filled with 10 g solid sulfur granules and 100 mL liquid medium only containing 0.05 g KNO\(_3\), 0.004 g KH\(_2\)PO\(_4\) and 0.2 g NaHCO\(_3\) (AR, Sinopharm Chemical Reagent Co., China). After 10-day culturing, aqueous samples collected from the supernatant were filtered through 20 μm membrane filter (diameter 18 cm, Nova, Hangzhou Special Paper Industry Co., China) to determine the nitrate concentration in the liquid medium. The NO\(_3\)\(^-\)-N removal efficiency was almost 100% during the culturing period (data not shown), demonstrated that the sulfur-utilizing autotrophic bacteria had been successfully acclimated.

2.3.2. Inoculation

According to different components for each stage as given in Table 1, the filter materials were adequately mixed by a shovel in a 25 L plastic bucket after washing. Firstly, the mixture of filter materials for ANS was packaged into the reactor; S/C layer (thickness 5 cm) filled in the middle of ANS, and 100 mL of the acclimated sulfur/liquid solution was inoculated onto the S/C layer. After the glass funnel was buried steadily, the mixture of filter materials was compacted firmly by shovel to reduce the porosity for creating an anaerobic environment. On the other hand, 100 mL of cultured nitrifying bacteria solution was taken, and then directly poured into a bucket, stirred by the shovel to fully mix.
with filter materials, and finally the mixture was filled into the upper part of the reactor to construct the AES.

2.3.3. Experimental procedure

The synthetic wastewater was stored in a 25 L plastic tank as reservoir and then pumped onto the surface sponge via a sprinkler at the flow rate of 20 mL min \(^{-1}\) by a peristaltic pump (Lead 15–24 Longer Precision Pump Co., China), and then flowed through AES and ANS by gravity, finally discharged from the bottom outlet. The hydraulic retention time (HRT) was 6 h. The SISSAD operated for 63 d at the room temperature (20–25 \(^{\circ}\)C). Samples were collected everyday from the influent, Effluent A and Effluent B.

2.4. Analytical methods

In this study, analytical reagent chemicals were used to prepare standards and stock solution (Sinopharm Chemical Reagent Co., China).

COD was digested by potassium dichromate using a 5B-1F Speed Digester (Lianhua Tech Co., China) at 150 °C for 10 min and determined by ultraviolet spectrophotometry at 610 nm using a spectrophotometer (Hach DR6000, USA). \(\text{NH}_4^+\)-N was measured by Nessler’s reagent colorimetry and determined using Hach DR6000 at 420 nm. TP was measured by molybdate colorimetry using Hach DR6000 at 700 nm, while \(\text{NO}_3^-, \text{NO}_2^-\) and \(\text{SO}_4^{2-}\) were analyzed using a Dionex ICS-2500 ion chromatograph (Dionex Corporation, Sunnyvale, USA). Water temperature and pH were monitored using a FE30 Benchtop Meter (Mettler Toledo. Co., USA).

3. Results and discussion

3.1. COD

It can be seen from Fig. 2, SISSAD showed an effective removal ability for COD, even on the first day, the COD concentration in Effluent A from AES sharply reduced from 462.75 to 229.76 mg L \(^{-1}\), by removal efficiency of 50.68%, and then COD gradually decreased. On Day 17, the COD concentration dropped to 40.82 mg L \(^{-1}\) with the removal efficiency of 91.95%, after then, the COD concentration varied at the range between 53.42 and 20.35 mg L \(^{-1}\), average COD removal efficiency was 88.44%. Therefore, it could be considered that the biofilming for AES was completed on Day 17. In comparison with former similar studies of Lei et al. (2013), whose biofilming period was 4 months, SISSAD shortened the biofilming period to nearly half a month, mainly due to the acclimation for nitrifying bacteria in this study. On the other hand, although COD was mostly consumed in AES, the COD concentration in Effluent B continued decreasing. Similar with Effluent A, on Day 1, the COD concentration in Effluent B decreased to 108.53 mg L \(^{-1}\) by the removal efficiency of 77.05%, on Day 15, reduced to the lowest concentration of 15.63 mg L \(^{-1}\), after that, the COD concentration was almost kept at a constant. It was showed that ANS also had the capacity for removing the residual COD from AES, the average COD concentration in Effluent B was 27.30 mg L \(^{-1}\). Therefore, SISSAD exhibited a high performance for removing COD, the average removal efficiency was 95.09%. In contrast, other soil treatment systems such as a SWI, the COD removal efficiency was 87.2% (Li et al., 2012) and a vertical flow CW was 88% (Abou-Elela and Hellal, 2012). The explanation for COD removal in this study was due to that (1) COD was consumed as nutrient for the growing of aerobes in AES; (2) COD was used as organic electron for heterotrophic denitrification in AES; (3) the activated carbon granules in the mixture maybe adsorbed a portion of COD (Zhao et al., 2013b) in both AES and ANS, especially in the initial period of the experiment.

3.2. \(\text{NH}_4^+\)-N

As shown in Fig. 3, on the first day of operating, the \(\text{NH}_4^+\)-N concentration in Effluent A from AES reduced to 22.08 mg L \(^{-1}\) with removal efficiency of 64.33%, and then the concentration continued to decrease. On Day 16, the \(\text{NH}_4^+\)-N concentration decreased to 4.14 mg L \(^{-1}\) by the removal efficiency of 92.39%. After then, the \(\text{NH}_4^+\)-N concentration was kept at less than 6 mg L \(^{-1}\), expect for some of them were over 6 mg L \(^{-1}\) due to higher influent \(\text{NH}_4^+\)-N concentration. Furthermore, the variation of \(\text{NH}_4^+\)-N concentration from Day 1 to Day 15 was almost consistent with COD variation in biofilming period mentioned above. However, the \(\text{NH}_4^+\)-N concen-

### Table 1

Component of filter materials.

<table>
<thead>
<tr>
<th>Stage</th>
<th>Quartz sand (S) (% v/v)</th>
<th>Quartz sand (L) (% v/v)</th>
<th>Volcanic (% v/v)</th>
<th>Zeolite (% v/v)</th>
<th>Activated carbon (% v/v)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AES</td>
<td>30</td>
<td>35</td>
<td>20</td>
<td>10</td>
<td>5</td>
</tr>
<tr>
<td>ANS</td>
<td>50</td>
<td>15</td>
<td>10</td>
<td>10</td>
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</table>
tration reached the inflection point two days earlier than COD, mainly due to that the ammonium adsorption by zeolite in the filter materials occurred. AES obtained a well ammonium removal capacity with the average NH$_4^+$-N removal efficiency of 89.99% in Effluent A.

On the other hand, the NH$_4^+$-N concentration in Effluent B from ANS on Day 1 declined to 5.52 mg L$^{-1}$ with the removal efficiency of 91.08%, the concentration continued to drop to 0.39 mg L$^{-1}$ on Day 12, obtaining the removal efficiency of 99.32%. After Day 12, the concentration of NH$_4^+$-N mostly remained below 5 mg L$^{-1}$, the average NH$_4^+$-N concentration in Effluent B was 2.86 mg L$^{-1}$ with the removal efficiency of 95.25%, meant that the residual NH$_4^+$-N from AES were used in ANS. The NH$_4^+$-N decrease in ANS was attributed to (1) adsorption by the filter materials; (2) utilization by sulfur-utilizing autotrophic denitrification according to Eq. (1). Seen from Eq. (1), 4 mol NH$_4^+$-N were needed for reducing 50 mol NO$_3^-$-N.

Therefore, the NH$_4^+$-N concentration requirement by autotrophic denitrification could be calculated as follow:

$$C_{\text{ANS}}(\text{NH}_4^+ - \text{N}) = \frac{4C_A(\text{NO}_3^- \text{-N}) \cdot E_{\text{ANS}}(\text{NO}_3^- \text{-N})}{50}$$

Furthermore, the practical NH$_4^+$-N concentration removed by ANS is:

$$C_{\text{ANS,pract}}(\text{NH}_4^+ - \text{N}) = C_A(\text{NH}_4^+ - \text{N}) - C_B(\text{NH}_4^+ - \text{N})$$

Where, $C_A$(NH$_4^+$-N) and $C_B$(NH$_4^+$-N) are the average NH$_4^+$-N concentrations of Effluent A and B (mg L$^{-1}$), $C_A$(NO$_3^-$-N) is the average NO$_3^-$-N concentration of Effluent A (mg L$^{-1}$), $E_{\text{ANS}}$(NO$_3^-$-N) is the average removal efficiency of ANS (%). Substituted 45.31 for $C_A$(NO$_3^-$-N), 6.02 for $C_A$(NH$_4^+$-N), 2.86 for $C_B$(NH$_4^+$-N) and 84.86 for $E_{\text{ANS}}$(NO$_3^-$-N), then $C_{\text{ANS}}$(NH$_4^+$-N) = 3.07 and $C_{\text{ANS,pract}}$(NH$_4^+$-N) = 3.16 were obtained. The practical NH$_4^+$-N removal was higher than the requirement by autotrophic denitrification, demonstrating that the additional adsorption process was involved.

Fig. 3. NH$_4^+$-N concentration and removal efficiency.
5250 g. As known, average influent NH$_4^+$-N concentration was 60.04 mg L$^{-1}$, and 64.33% NH$_4^+$-N removal in the reactor on Day 1 was hypothesized as the adsorption efficiency, 772.47 mg NH$_4^+$-N could be adsorbed by the zeolite in the reactor every day (20 L synthetic wastewater). However, 5250 g zeolite could adsorb 21721.88 mg NH$_4^+$-N calculated using the HRT 6 h, then divided by 772.47 mg d$^{-1}$, figuring out 28 days as the theoretical saturated adsorption period for NH$_4^+$-N, and it was close to 24 days mentioned above, after which the ammonia removal was considered resulting from microbial activities. Nevertheless, owing to the oxidation of NH$_4^+$-N, zeolite might never achieve a practical saturated adsorption for NH$_4^+$-N. Thus, the NH$_4^+$-N removal was due to both adsorption and nitrification.

After Day 15, the biofilm of nitrifying bacteria was gradually formed, the conversion from ammonium to nitrite occurred by nitrifying bacteria became a dominant role instead of adsorption.

Overall, the average NH$_4^+$-N removal efficiency of the whole system was 95.25%, and the highest removal efficiency was 99.32%, exhibiting that the SISSAD had a high performance on NH$_4^+$-N removal, also evidenced that artificial aeration could enhance ammonia removal in soil infiltration system.

### 3.3. NO$_3^-$-N and NO$_2^-$-N

As shown in Fig. 4, the concentration of NO$_3^-$-N in Effluent A was 3.57 mg L$^{-1}$ on Day 1, and then increased rapidly. After 7 days, the NO$_3^-$-N concentration in Effluent A had already increased to 43.67 mg L$^{-1}$, and then continued to grow up to the highest concentration of 62.55 mg L$^{-1}$ until Day 25. Since Day 26, the concentration had been slightly floating for 22 days. Adversely, the NO$_3^-$-N concentration tended to decrease slowly after Day 49, finally ending with 45.87 mg L$^{-1}$.

The NO$_3^-$-N concentration in Effluent A was 0.09 mg L$^{-1}$ on Day 0, and smoothly increased to 0.81 mg L$^{-1}$ on Day 17 with a small fluctuation, then rapidly growing to 2.50 mg L$^{-1}$ on Day 26. After then, the concentration began to slightly decrease and float by a tiny range between 1.30 and 2.49 mg L$^{-1}$, owning an average concentration of 1.40 mg L$^{-1}$.

As discussed above, the average NH$_4^+$-N removal efficiency in AES was 89.99%, it was hypothesized that the 89.99% of NH$_4^+$-N were all converted into NO$_3^-$-N, a theoretical average NO$_3^-$-N concentration of 60.04 mg L$^{-1}$ would be obtained from AES. However, the average NO$_3^-$-N concentration detected in Effluent A was 45.31 mg L$^{-1}$. The disagreement between theoretical and practical concentration was probably due to: (1) a portion of NH$_4^+$-N was used as nutrient substance for the growth of microorganisms to produce biomass such as cell wall; (2) denitrification might be taken place in some anoxic parts of AES, consequently leads to the formation of NO$_2^-$-N. Yang et al. (2013b) also reported that the reduction of NO$_3^-$-N in anoxic denitrification would bring NO$_2^-$-N into existence. On the other hand, NO$_3^-$-N was found to be an obligate intermediate in nitrification (Morkved et al., 2007), and previous researches on CW indicated that most conventional subsurface flow constructed wetlands were unable to fulfill the process of NH$_4^+$-N converting into NO$_3^-$-N on occasion of lack of oxygen, resulting in nitrite generation (Fan et al., 2013). Therefore, it was indicated that some portions in AES remained in an anoxic condition due to that non-uniform oxygen supplying resulted from a single pipe in this study.

The NO$_3^-$-N concentration in Effluent B was kept at the range from 0.40 mg L$^{-1}$ to 9.33 mg L$^{-1}$. The average NO$_3^-$-N concentration in ANS was 4.35 mg L$^{-1}$ with the removal efficiency of 84.86%, demonstrating a high denitrification performance in the ANS.

Seen from Eq. (1), autotrophic denitrification needed inorganic carbon (CO$_2$ or HCO$_3^-$), however, ANS still gave a well denitrification performance even though inorganic carbon was not added in the synthetic wastewater. A previous study has revealed a mixotrophic process combining simultaneous heterotrophic denitrification with autotrophic denitrification in sulfur-based column bioreactor (Sahinkaya and Dursun, 2012). The explanation in this study might be:

1. CO$_2$ generated from aerobic respiration by heterotrophic aerobes, following Eq. (4):

\[ C_6H_{12}O_6 + 6O_2 \rightarrow 6CO_2 + 6H_2O \quad \text{(4)} \]

A stoichiometric calculation based on aerobic respiration for glucose indicated that consuming 88.44% of COD (average influent concentration 500.32 mg L$^{-1}$) in AES would produce 608.41 mg L$^{-1}$ CO$_2$.

2. CO$_2$ generated from the denitrification in some anoxic parts in AES, following Eq. (5):

\[ C_6H_{12}O_6 + 12NO_3^- \rightarrow 6CO_2 + 6H_2O + 12NO_2^- \quad \text{(5)} \]

Therefore, 0.73 mg L$^{-1}$ CO$_2$ and 1.40 mg L$^{-1}$ NO$_2^-$-N would be generated.

3. CO$_2$ from the reaction of SO$_4^{2-}$ with CaCO$_3$. In fact, this portion of CO$_2$ might not be the main source of inorganic carbon, but CO$_2$ from acidized CaCO$_3$ also played a role in supplying additional inorganic carbon source.

![Fig. 4. NO$_3^-$-N and NO$_2^-$-N concentration.](image-url)
However, according to Eq. (1), to reduce 45.31 mg L\(^{-1}\) NO\(_3\)-N from Effluent A, only 56.96 mg L\(^{-1}\) CO\(_2\) was needed, indicated that CO\(_2\) from aerobic respiration by heterotrophic bacteria was enough to supply inorganic carbon source for sulfur-utilizing autotrophic denitrification.

On the other hand, NO\(_3\)-N occurred in AES was mostly removed by ANS with sulfur/limestone system, obtaining an average NO\(_3\)-N concentration of 0.18 mg L\(^{-1}\) and an average removal efficiency of 87.41% in ANS, because sulfur-based autotrophic denitrification could reduce nitrite to N\(_2\) as well as nitrate (Qambrani et al., 2013).

### 3.4. TP

As shown in Fig. 5, total phosphorus (TP) in Effluent A decreased from 10.11 mg L\(^{-1}\) to 5.32 mg L\(^{-1}\) on the first day, and then fluctuated from 2.01 mg L\(^{-1}\) to 6.22 mg L\(^{-1}\) until Day 18. After Day 19, the TP concentration began to stabilize and remained stable around 3 mg L\(^{-1}\). The average TP concentration from AES was 3.13 mg L\(^{-1}\) with the removal efficiency of 68.39%, mainly due to the adsorption and consumption as nutrient by microorganisms.

Flowing through ANS, TP in Effluent B gradually decreased from 2.02 mg L\(^{-1}\) on Day 1 to 0.92 mg L\(^{-1}\) on Day 20, and then the TP concentration had never been up to 1 mg L\(^{-1}\). The average TP concentration in ANS was 0.67 mg L\(^{-1}\) and the total removal efficiency in the reactor was 93.15%, which was superior to 60% of bioretention systems (Ergas et al., 2010). In this study, it was considered that the volcanics in the mixed-filter-materials played a significant role in adsorbing phosphorus. Yang et al. (2013a) have reported that volcanics such as Kunama mud exhibited great phosphate adsorption capacity. Furthermore, phosphate was necessary for the process of autotrophic denitrification by denitrifying bacteria because phosphorus was used by denitrifying bacteria for producing ADP sulfurylase (Bruser et al., 2000), consequently phosphate could be removed.

### 3.5. SO\(_4^{2-}\) and alkalinity

As shown in Fig. 6, the growing of SO\(_4^{2-}\) indicated that denitrifying bacteria for autotrophic denitrification used sulfur granules in the ANS. The SO\(_4^{2-}\) concentration on the first day was only 28.46 mg L\(^{-1}\), and then smoothly rose up to 166.93 mg L\(^{-1}\) by Day 49. However, after then, the SO\(_4^{2-}\) concentration rapidly increased, reaching 396.22 mg L\(^{-1}\) on the last day.

During the experiment period, the pH value in Effluent A floated from 7.36 to 6.37 due to the oxidation of NH\(_4\)-N and the generation of nitrate and nitrite, which accorded with Yuan’s report on soil wastewater infiltration system (Yuan et al., 2013). Nevertheless, pH in Effluent B decreased from 6.8 to 5.32, much lower than Effluent A, suggested that sulfur-utilizing autotrophic denitrification in ANS had more hydrogen ions generated than that in AES. After Day 52, the pH in Effluent B sharply dropped to 5.24, at which the internal environment was no longer suitable for autotrophic denitrifying bacteria. It has been reported that sulfur-utilizing autotrophic denitrification occurred between pH 6 and 9 (Oh et al., 2001), and SO\(_4^{2-}\) showed severe inhibition on denitrification process at pH lower than 5.5 (Liu and Koeing, 2002). Unfortunately, pH continued to lower down, finally a small amount of solid particle impurities with dark color was observed and sour smelled in the final effluent. The impurity was speculated to be a mixture containing dead bodies of autotrophic anaerobes and the acidized limestone particles. The SISSAD was sacrificed immediately on Day 63 when pH tragically dropped to 3.84. Compared with Ergas’ 76 cm-high biofilter for 3 months (Ergas et al., 2010), the practical operating period of SISSAD seemed rather short, which yet does not mean a vertical infiltration system is not as superior as those up-flow sulfur packed-bed reactors (USPR) or a horizontal-flow bioreactor (Read-daily et al., 2011) since the excess and uncontrollable consumption of alkalinity was not estimated. The alkalinity reducing caused by a growing sulfate concentration was inevitable in sulfur/limestone autotrophic denitrification (SLAD) (Moon et al., 2006; Zhou et al., 2011), however, in contrast to USPRs, SISSAD had more alkalinity consumption mainly due to the restriction of vertical water flow. Former issues on CW suggested that vertical-flow had greater oxygen transfer capacity than others (Cui et al., 2012). Similarly, in this study, owing to the vertical-flow infiltration design, a non-uniform water distribution caused the increasing of DO and much quicker neutralizing reaction occurred in some parts in the mixed-filter-materials, resulting more alkalinity consuming. Moreover, low hydraulic loading rate (HLR) and long artificial aeration time could bring about an excellent nitrogen removal, while also led the sulfur oxidized rapidly. To avoid the risk of harming aquatic environment, SO\(_4^{2-}\) in the final effluent would be further treated by some approaches such as CaCO\(_3\) filter process in the practical application.

A stoichiometric calculation was performed based on Eq. (1) to explain the fatal problem involved with alkalinity. We hypothesized that all NH\(_4\)-N from the synthetic wastewater were converted into NO\(_3\)-N and then reduced into N\(_2\) by sulfur-utilizing denitrifying bacteria, means that removing an average 60.04 mg L\(^{-1}\) NO\(_3\)-N will generate 457.87 mg L\(^{-1}\) SO\(_4^{2-}\) and
consume 274.47 mg L\(^{-1}\) CaCO\(_3\) as alkalinity. However, the average SO\(_4^{2-}\) concentration in effluent was only 149.59 mg L\(^{-1}\) and highest SO\(_4^{2-}\) concentration was 396.22 mg L\(^{-1}\), demonstrated that crushed oyster shells were effective on removing most of SO\(_4^{2-}\) and neutralizing H\(^+\). The rapid increasing of SO\(_4^{2-}\) after Day 56 (Fig. 6) indicated that oyster shells were no longer able to maintain the pH over 6 for an adequate autotrophic denitrification. According to stoichiometric calculation, the total alkalinity consumption for the whole experiment period was 354.83 g CaCO\(_3\) (20 L synthetic wastewater per day), however, 513.42 g of the prepared oyster shell only survived for 2 months. This might be due to that (1) the large cross sectional area for the SISSAD reactor under low HLR may cause a non-uniform water distribution, consequently resulted in lower utilization of oyster shells; (2) large particle size of crushed oyster shells could also result in low utilization. It was suggested dispersing the S/C particles instead of gathering them in a layer, and reducing the particle size of oyster shells would be necessary in the future study. On the other hand, a volume ratio of 3:1 for S/C was considered no longer fit for the sulfur-utilizing autotrophic denitrification but gaining fewer sulfates (Li et al., 2013a).

4. Conclusion

The lab-scale SISSAD, combining soil infiltration with SLAD had been constructed. Except for its uncontrollable sulfate generation, the system had shown a remarkable performance on treating synthetic domestic wastewater. The majority of COD and NH\(_4^+\)-N were removed in AES by aerobic respiration and nitrification respectively; phosphorus was mostly removed by adsorption, while nitrate and nitrite were reduced by autotrophic denitrification in ANS. The integration design brought about a compact construction that allows CO\(_2\) generated from AES to be directly used in ANS, consequently lowering cost and space occupation. Further study will be focused on oxygen supply and alkalinity consumption.

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