Performance of nitrogen removal in attached growth reactors with different carriers

H. T. Le, N. Jantarat, W. Khanitchaidecha, K. Ratananikom and A. Nakaruk

ABSTRACT

Two waste materials, concrete and sponge, were used as biomass carriers in the attached growth reactor in a nitrogen wastewater treatment system. The nitrogen removal performance was compared to a control reactor using commercial carrier material. The highest nitrogen removal efficiency, 87%, was found in the sponge reactor, with the concrete reactor showing 82% efficiency ahead of the commercial reactor of 76%. A thick biofilm developed on the fiber of the sponge carrier, with the biomass increasing from 270 g-VSS/m^3-carrier to 1,000 g-VSS/m^3-carrier. For the concrete carrier, biomass was observed on the concrete cracks and also as a biofilm on the surface. The maximal biomass was 630 g-VSS/m^3-carrier. The content of the biomass agglomerated in the commercial carrier was 310 g-VSS/m^3-carrier. Nitrification and denitrification simultaneously occurred to remove nitrogen in the sponge and the commercial carrier reactor. However, in the concrete reactor, nitrification mainly occurred during the aeration phase and denitrification occurred in the non-aeration phase. These results demonstrate that the sponge was the best carrier, with high nitrogen removal efficiency, dense biomass and tolerance to shock loading. The simplicity inherent in the system design together with good performance make it suitable for use in wastewater treatment systems.

Key words | attached growth reactor, commercial carrier, concrete carrier, sponge carrier, wastewater treatment

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INTRODUCTION

The removal of nitrogen, including ammonium-nitrogen (NH_4-N), nitrite-nitrogen (NO_2-N) and nitrate-nitrogen (NO_3-N), from domestic wastewater is an important issue for water resource management, especially for controlling polluting water runoff with a high nitrogen content. High nitrogen levels in wastewater discharged into local water sources, such as ponds and small catchments, common in rural areas, can often result in toxic blooms of algae and 'scum' on water surfaces, rendering it unfit for fish breeding, crop watering and animal consumption, important uses in rural areas. Domestic wastewater usually has high nitrogen content and is an easily identifiable source as well as being easily treated at the local level.

Several physical and biological treatment methods have been developed to remove the nitrogen from domestic wastewater. These treatment methods include granular nitritation and anammox in membrane-aerated biofilm reactors (Li *et al.* 2016), hybrids of activated carbon and sequencing batch reactors (Sirianuntapipoon & Chairattanawan 2016), and combined subsurface and surface flow constructed wetlands (Sartori *et al.* 2016). Current treatment systems are mainly focused on the modification of exiting traditional systems to enhance their treatment ability with co-removal mechanisms. However, more importantly in remote areas, a reliable supply of clean water is an imperative, and therefore reliable domestic wastewater treatment plants of simple

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design, using uncomplicated materials available locally, with low operating costs, to remove the nitrogen from the wastewater, are essential. The attached growth reactor is a type of biological treatment plant, in which the active sludge is attached to a carrier as a biofilm, is such a reliable technology appropriate to remote areas.

The positive effects on the nitrogen removal performance of biomass immobilization in the attached growth reactor have been discussed previously (Xiangli *et al.* 2008; Dong *et al.* 2016). The advantages of these attached growth reactors included high biomass concentration, simultaneous nitrification and denitrification, and resistance to nitrogen shock loading and other environmental changes. In recent years, various natural and artificial solid materials have been proposed as the carrier in attached growth reactors for wastewater treatment. Natural zeolite and activated carbon have been used as the biomass carrier (Park *et al.* 2003; Feng *et al.* 2014) which had the significant advantage of the co-removal mechanisms of adsorption and biological processes. Well-known artificial carriers are made from polyethylene and polyurethane. The polyurethane form carrier can improve the efficiency of long-term nitrification and denitrification and sustain microbial populations of slow growing microorganisms (Daniel *et al.* 2009). Biodegradable polymers (such as polycaprolactone) have also been used in treating domestic wastewater. A biodegradable carrier can provide the soluble organic carbon for denitrification in wastewater containing a low C/N ratio (Chu & Wang 2011).

Although there has previously been extensive research on nitrogen removal performance, and microbial communities, in attached growth reactors, with a variety of carriers, the use of waste material as the biomass carrier for biomass attachment to enhance the nitrification and denitrification has not been comprehensively explored. Concrete is a construction waste product which is mostly disposed by open dumping in landfills or otherwise illegally dumped. The surface roughness and high voids of concrete make it potentially useful as a biomass carrier. Another waste product, sponge, probably used in all households in the nation, is a porous material appropriate for biomass attachment, inexpensive and readily available, even in remote areas.

Prior to this, carbon in various forms has been used for nitrogen removal from domestic wastewater. This is because the wastewater has a low carbon content which is insufficient for complete denitrification. In previous studies, the carbon concentration was maintained in the range of 500–600 mg/L, and the C/N ratio was around 25 (Feng *et al.* 2008; Liu *et al.* 2010). Although excellent performance of nitrogen removal has been attained in previous studies (Walters *et al.* 2009), the residual carbon can impact the quality of the treated water. Further, when the biodegradable material acted as carrier and carbon source, the amount of carbon release and stable release rate were matters of concern.

Therefore, the aim of this study was to investigate the possibility of using alternative materials for domestic wastewater treatment in the attached growth reactor. Concrete and sponge were considered, for the reasons previously stated. The nitrogen removal performance at low carbon content levels of an attached growth reactor, using variously the concrete and the sponge material as the carrier, was investigated. The performance of these materials was compared to that of a commercial carrier which is commonly used in attached growth reactor wastewater treatment plants. The removal mechanisms occurred in the different carriers was clarified, and the attached biomass was observed for volume and time of growth.

METHODOLOGY

Carrier

Concrete and sponge are two waste materials which were used for biomass attachment in this research. The concrete which came from a construction site and sponges from a local market were cut into 2 cm cubic shape. The commercial carrier with 2 cm diameter was used as a control to compare the performance. The carriers were transferred to a suspended sludge which had been cultivated by NH_4-N feeding for a month. The accumulated biomass was in the range of 270 g-VSS/m^3-carrier for the concrete and sponge carriers and 180 g-VSS/m^3-carrier for the commercial carrier.

Experimental set-up

Three attached growth reactors were established (Figures 1 and 2). The size of each reactor was 11 cm internal diameter and 40 cm height with a working volume of about 3 L. The

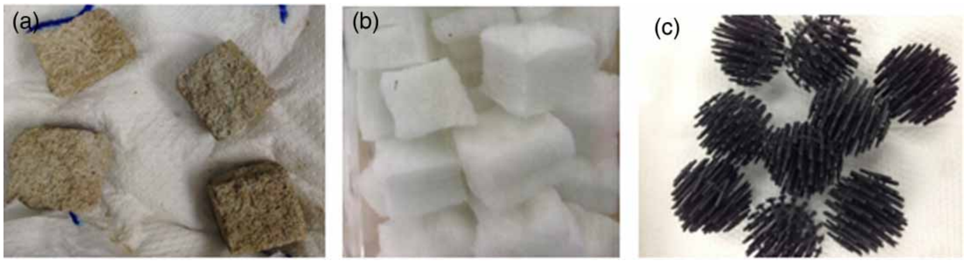

Figure 1 | (a) Concrete carrier, (b) sponge carrier and (c) commercial carrier.

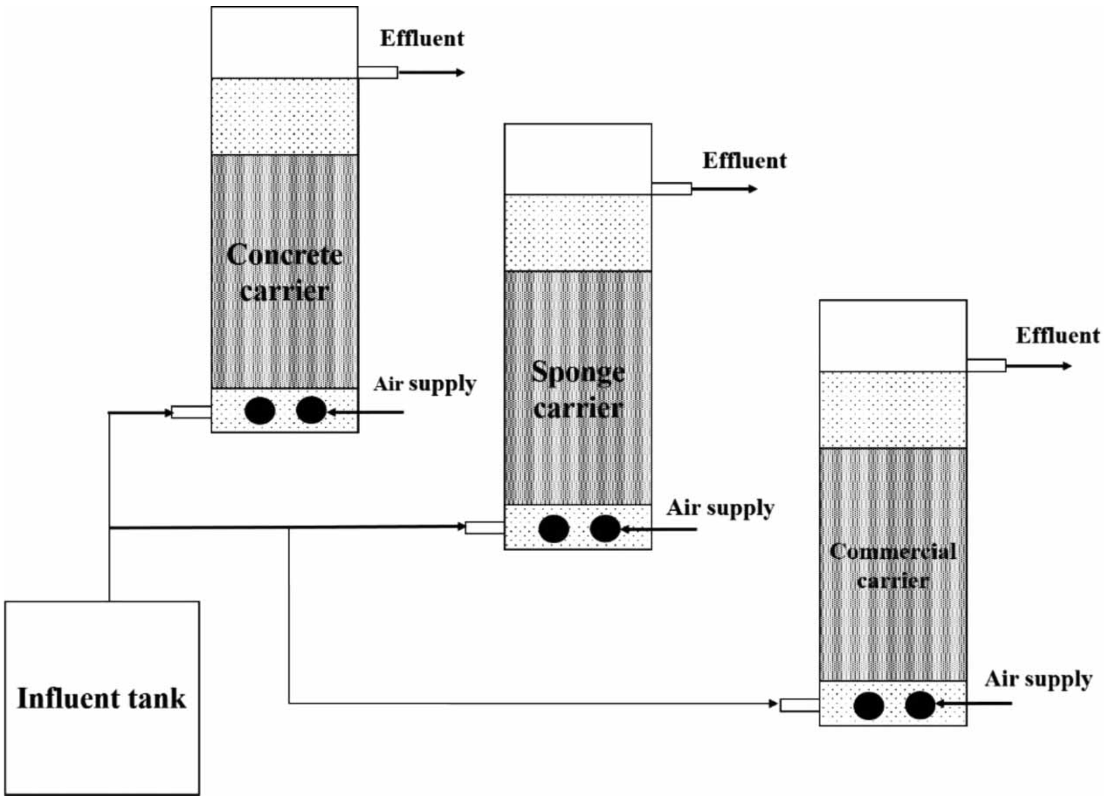

Figure 2 | Schematic of attached growth reactors.

three carriers – concrete, sponge and commercial – were each added to a reactor to be 60% of the working volume. The synthetic wastewater (influent) was continuously fed to the reactors at the flow rate of 5 L/day. The reactors had a hydraulic retention time of 12 hours. Air was intermittently diffused through an air stone in a three-hour aeration and five-hour non-aeration cycle. The continuous influent flow along the reactors height enhanced the mass transfer and water circulation during the aeration and non-aeration cycle. The aeration period enabled nitrification and the non-aeration (anaerobic) period allowed denitrification in a manner suggested in Le *et al.* (2015). The dissolved oxygen (DO) was around 5 mg/L in the aeration period and was gradually decreased to 0.5 mg/L in the first hour of the anaerobic period.

In the experiment, there were three conditions with different influent NH_4-N and organic carbon concentrations. An initial concentration of 40 mg/L of NH_4-N was fed to the reactors in Phases 1 and 2, and the NH_4-N concentration was then increased to 80 mg/L in Phase 3.

The C/N ratio was kept at 2.5 in Phase 1 and increased to 3.5 in the sequencing phases (in Table 1).

Synthetic wastewater preparation

Synthetic domestic wastewater was used for evaluating the reactor performance. This wastewater was prepared by mixing in the following chemicals (g/L); NH_4Cl 0.15–0.30, CH_3COONa 0.34–0.48, KH_2PO_3 0.02, $MgSO_4$ 0.03, $CaCl_2$ 0.36, $FeSO_4$ 0.003 and trace elements 0.5 mL (Guo *et al.* 2013). The concentrations of NO_2-N and NO_3-N were lower than 1 mg/L while the NH_4-N concentration was between 40 mg/L and 80 mg/L.

Biomass measurement

The carriers were taken from the attached growth reactors at the initial stage, day 0, and day 45 and 70 (the steady state day). The biomass was removed from the carriers by rinsing and shaking for 15 minutes, and the liquid was then centrifuged and filtered. The mixed liquor suspended solids and mixed liquor volatile suspended solids concentrations in the biomass samples were measured according to the standard method (APHA 1998). The biomass content was calculated as g-VSS/m^3-carrier. In addition, the carrier and its biofilm were observed by using a scanning electron microscope (Leo 1400 Series).

Analytical method

The performance of the attached growth reactors was measured by two parameters: NH_4-N removal efficiency and nitrogen removal efficiency. The reduction of NH_4-N between the influent and effluent was only concerned with the NH_4-N removal efficiency which refers to nitrification ability. However, all nitrogen forms of NH_4-N, NO_2-N and NO_3-N were included in the nitrogen removal efficiency which refers to both nitrification and denitrification. The influent samples were immediately taken from the synthetic wastewater after preparation, while the effluent samples were collected from the end of non-aeration (after 24-hour operation). The concentrations of NH_4-N, NO_2-N and NO_3-N were analyzed in accordance with the standard method (APHA 1998). NH_4-N removal efficiency and nitrogen removal efficiency were calculated according to Equations (1) and (2). The specific nitrogen removal which referred to the nitrogen removal per biomass unit was calculated according to Equation (3). The organic carbon was measured using a total organic carbon analyzer (HACH, IL530 TOC-TN). The pH and DO were measured daily using a pH meter (Eutech Instruments) and a DO meter (CyberScan DO 110 Model).

$$\text{NH}_4\text{-N removal efficiency (\%)} = \left(1 - \frac{NH_{4\,\text{effluent}}}{NH_{4\,\text{influent}}}\right) \times 100 \tag{1}$$

$$\text{Nitrogen removal efficiency (\%)} = \left(1 - \frac{NH_{4\,\text{effluent}} + NO_{2\,\text{effluent}} + NO_{3\,\text{effluent}}}{NH_{4\,\text{influent}} + NO_{2\,\text{influent}} + NO_{3\,\text{influent}}}\right) \times 100 \tag{2}$$

$$\text{Specific nitrogen removal(mg N/g VSS)} = \left(\frac{\text{Total N in influent} - \text{Total N in effluent}}{\text{Biomass}}\right) \tag{3}$$

Table 1 | Operating condition of attached growth reactors

Carrier	Influent NH_4-N (mg N/L)	Organic carbon (mg C/L)	C/N	Operation day
Concrete	40	100	2.5	Phase 1: day 1–20
	40	140	3.5	Phase 2: day 21–45
	80	280	3.5	Phase 3: day 46–70
Sponge	40	100	2.5	Phase 1: day 1–20
	40	140	3.5	Phase 2: day 21–45
	80	280	3.5	Phase 3: day 46–70
Commercial carrier	40	100	2.5	Phase 1: day 1–20
	40	140	3.5	Phase 2: day 21–45
	80	280	3.5	Phase 3: day 46–70

RESULTS AND DISCUSSION

Performance of attached growth reactors

At the low NH_4-N loading in Phase 1, the nitrogen removal efficiency for the three attached growth reactors increased continuously until it became stable at the steady state on day 45. In the steady state, the sponge reactor showed the highest nitrogen removal efficiency of 68%, followed by the concrete reactor of 61% and the commercial carrier reactor of 56% (Figure 3). However, the NH_4-N removal

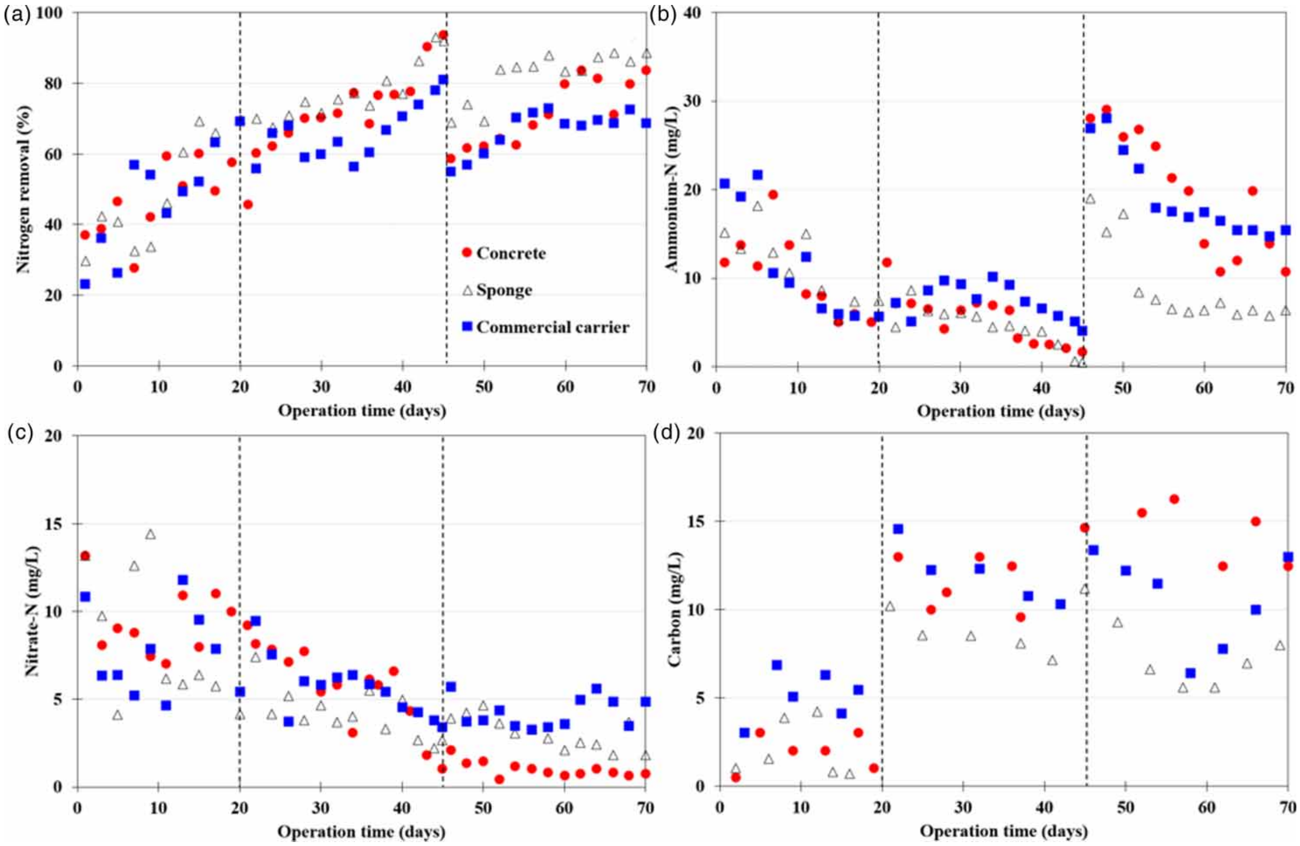

Figure 3 | (a) Nitrogen removal efficiency, (b) effluent NH_4-N concentration, (c) effluent NO_3-N concentration and (d) effluent organic concentration.

efficiency for all three reactors was 85% (data not shown). These results show that nitrification is more effective than denitrification for oxidizing NH_4-N to NO_3-N. The low concentration of organic carbon in the effluent indicated that the nitrification and denitrification was limited by the rate of denitrification due to the inadequate supply of carbon.

In Phase 2, the nitrogen removal efficiency continued to increase at the higher C/N ratio of 3.5. At the steady state, the nitrogen removal efficiency achieved 82% for the concrete reactor, 87% for the sponge reactor and 76% for the commercial carrier reactor. Similarly, the NH_4-N removal efficiency also increased and reached 90–95% for all reactors. The dramatic increase in nitrogen removal efficiency was due to there being sufficient carbon available for microbial metabolism to remove the nitrogen, as indicated by there being residual carbon in the effluent. In addition, the growth of the biomass on the carriers was another significant reason for the dramatic increase in nitrogen removal efficiency. The biomass content increased from 270 g-VSS/m^3-carrier in the initial stage to 610 and 1,000 g-VSS/m^3-carrier for the concrete and sponge at the steady state, and increased from 180 g-VSS/m^3-carrier to 290 g-VSS/m^3-carrier for the commercial carrier (Figure 4). It can be seen that dense biomass was observed in the sponge and concrete carriers, which has a high porosity and particular surface characteristics which encouraged the faster attachment of biomass, whereas the commercial carrier has a smooth and rigid surface not as conducive to biomass attachment.

At the high NH_4-N loading of 80 mg/L in Phase 3, the nitrogen removal efficiency suddenly dropped and then increased again to 80% for the concrete reactor, 88% for the sponge reactor and 70% for the commercial carrier reactor. The nitrogen removal efficiency of each carrier at steady state showed that the nitrogen removal efficiency values at the high NH_4-N loading were very similar to those at the low NH_4-N loading. This means that all three reactors can effectively operate at either low or high NH_4-N loadings. In addition, the biomass content slightly increased to 630, 1,030

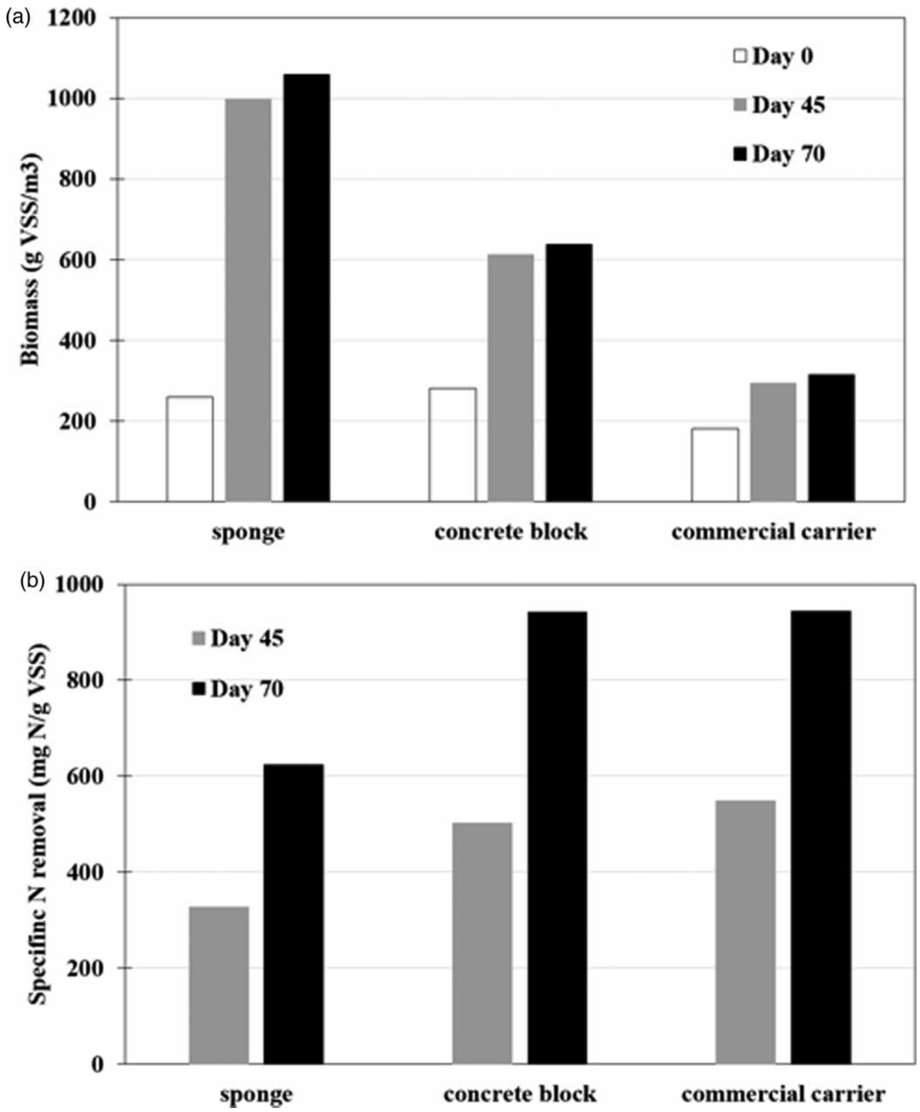

Figure 4 | (a) Biomass content and (b) specific nitrogen removal on various carriers.

and 310 g-VSS/m^3-carrier for the concrete, sponge and commercial carriers respectively (Figure 4(a)). Figure 4(b) shows that the specific nitrogen removal from the concrete and commercial reactors had a similar value of 940 mg-N/g-VSS, but the value was only 620 mg-N/g-VSS for the sponge reactor. This indicates that the sponge reactor can efficiently treat wastewater containing higher NH_4-N (>80 mg/L) up to the point when the specific nitrogen removal value reached the maximum of approximately 940 mg-N/g-VSS.

The results clearly demonstrate that the waste materials of concrete and sponge can be used as effective and efficient biomass carriers in a biological nitrogen treatment system. The effectiveness of nitrogen removal was in the following order: sponge > concrete > commercial carrier. This variation in nitrogen removal which was related to the porosity and surface roughness. Moreover, the sponge carrier shows tolerance to high NH_4-N loading and retains excellent performance.

Characteristics of biomass

During operation, the biomass developed as a biofilm covering the carrier. The biofilm on the sponge carrier grew rapidly from 270 g-VSS/m^3-carrier at the initial state to 1,000 g-VSS/m^3-carrier in 45 days. The biofilm on the commercial carrier grew more slowly due to the biofilm falling off the smooth surface of the commercial carrier. The sponge carrier was made from cellulose wood fiber which provided the larger surface area for biomass attachment and avoided biofilm scouring. In contrast, the commercial carrier was made from plastic and contained large cavities which negatively impacted on the biomass growth and strength of attachment. The concrete carrier had a rough surface, which was appropriate for biomass attachment, but the rigid structure of the material prevented the growth of biomass inside the concrete.

Figure 5(a) shows the surface of the concrete carrier with the biomass covering the surface as a biofilm and also in the rough indents or cracks on the concrete surface. For the sponge carrier, the biomass was mostly attached on the fiber as a biofilm; a thick biofilm was observed on the surface and on the fibers in the core of the sponge carrier. The biofilm was only slightly developed on the commercial carrier and had agglomerated as microbial floc on its internal structures.

Nitrogen removal model of carriers

At the end of the experiment (day 70), the mechanisms of nitrogen removal in three attached growth reactors was clarified. The reactors were operated under intermittent air

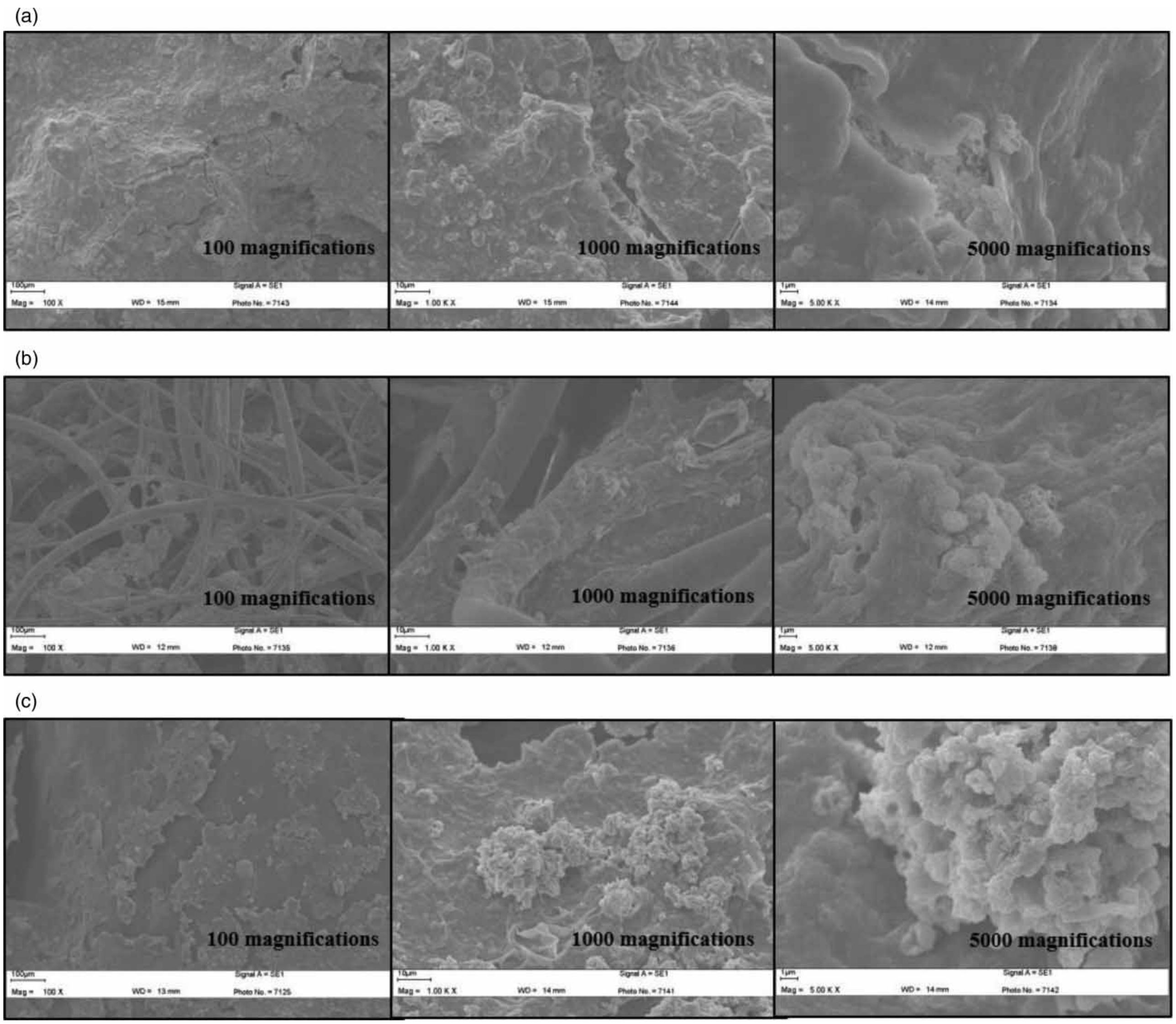

Figure 5 | Scanning electron microscope images of (a) concrete carrier, (b) sponge carrier, and (c) commercial carrier.

supply (three hours of aeration and then five hours of non-aeration), and the change of nitrogen levels in the eight-hour cycle was demonstrated. The level of NH_4-N in the concrete reactor continuously decreased in the three hours of aeration, while the NO_3-N was increasing. When the non-aeration phase started at hour 4, the NO_3-N immediately decreased and reached the low level of 0.5 mg/L (Figure 6). These results suggest that the nitrification to oxidize NH_4-N to NO_3-N firstly occurred in the aeration phase, and the denitrification which occurred in the non-aeration phase reduced the NO_3-N to N_2 gas.

In term of the biomass attached to the concrete carrier, the biofilm of biomass fully covered the concrete surface and no biomass was detected in the core. The biomass required oxygen to encourage nitrification, but the high oxygen during the aeration phase inhibited the denitrification from occurring simultaneously. The concrete carrier required the non-aeration phase, in which the very low oxygen was observed for denitrification to occur as presented in Figure 6(a). Nitrification was possible during oxygen depletion in the non-aeration phase. However, the increasing NH_4-N in the non-aeration phase was due to the continuous influent feeding, with NH_4-N accumulating in the reactor. The NH_4-N accumulation had more effect on the actual concentration than the decrease in NH_4-N from nitrification. Nitrifiers and denitrifiers existed concurrently on the carrier surface, and each microorganism played an important role as the key microbial activity in appropriate environment.

For the sponge carrier, the concentration of NH_4-N, NO_2-N and NO_3-N changed slightly in the eight-hour cycle. This is because the nitrification and denitrification can simultaneously occur within the biofilm, in which there are DO gradients because of diffusion limitation (Kotlar *et al.* 1996). Nitrification took place at the aerobic layer of the sponge surface, whereas denitrification happened in the deeper layer of biofilm which was an anoxic zone. Further, the NH_4-N concentration was stable during the non-aeration possibly because the nitrifier in the sponge carrier can metabolize under either low or high oxygen (Fitzgerald *et al.* 2015).

Similarly, simultaneous nitrification and denitrification was found in the commercial carrier, as indicated by slightly changing NH_4-N and NO_3-N concentrations. As discussed above, the high oxygen on the commercial carrier surface induced nitrification to occur in the outer microbial floc, and the low oxygen from diffusion resulted in denitrification in the inner floc. From all results, the concrete and sponge carriers can perform well in removing nitrogen from wastewater; however, the mechanisms of nitrogen removal were

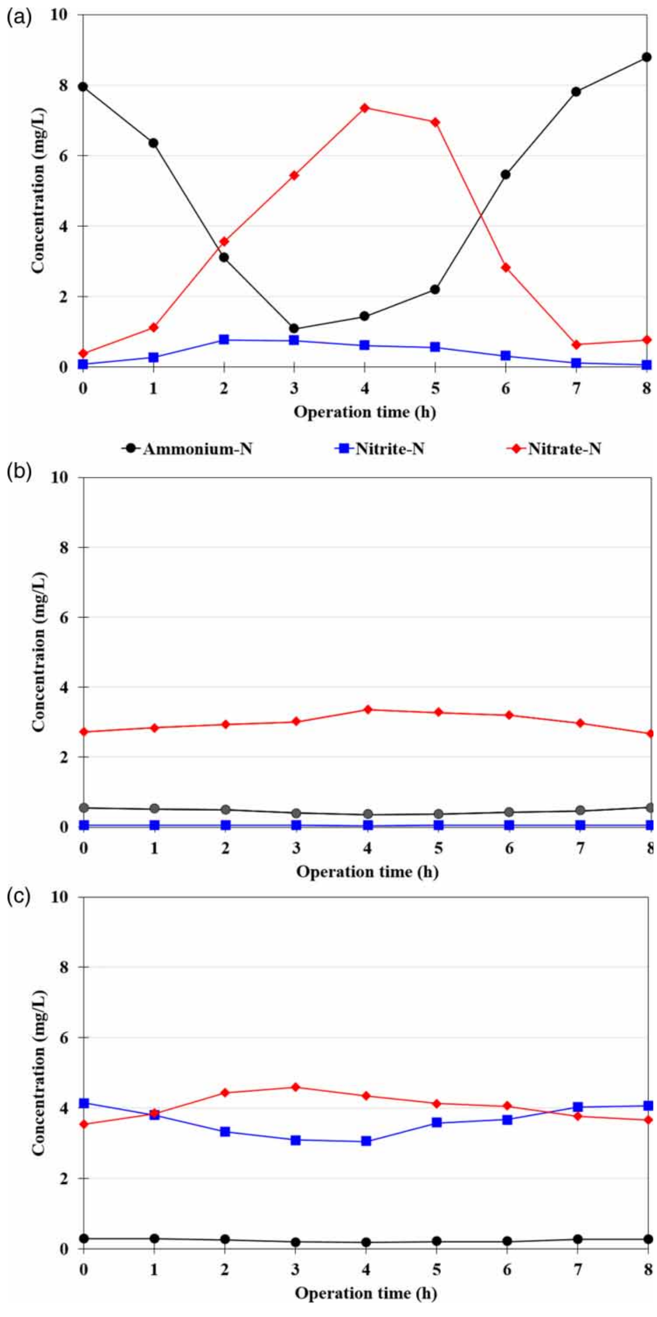

Figure 6 | Change of NH_4-N, NO_2-N and NO_3-N in (a) concrete reactor, (b) sponge reactor, (c) commercial carrier reactor.

significantly different which affected the operating conditions required to achieve effective reactor performance.

CONCLUSIONS

Concrete and sponge waste materials can be used as the biomass carrier in nitrogen wastewater treatment. The performance of both in attached growth reactors was better than the use of a commercial carrier which is commonly found in wastewater treatment system. The highest nitrogen removal efficiency of 87% was found in the sponge reactor: this is due to the high biomass attachment of 1,030 g-VVS/m^3-carrier. The efficiencies of concrete and commercial carrier reactors were 82% and 76% respectively. The porosity and surface roughness of the carriers impacted on biomass attachment and mechanism of nitrogen removal. The biomass of 630 g-VSS/m^3-carrier was mainly found on the surface of the concrete carrier, thus the concrete reactor requires intermittent aeration to obtain nitrification and denitrification. On the other hand, the biomass attached in the inner cavities of the sponge and agglomerated as microbial floc in the commercial carrier, therefore simultaneous nitrification and denitrification can occur without a non-aeration period.

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