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High-rate nitrogen removal and its behavior of granular sequence batch reactor under step-feed operational strategy

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HIGHLIGHTS

- High nitrogen removal efficiency was achieved in step-feed granular SBR.
- Higher nitrogen removal rate was obtained in granular SBR under step-feed mode.
- Granules were well maintained under the step-feed operational strategy.

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ABSTRACT

Alternating anoxic/oxic (A/O) combined with the step-feed granular sequence batch reactor (step-feed SBR) was operated in laboratory scale to investigate nitrogen removal. The results showed that when the total inorganic nitrogen (TIN) and chemical oxygen demand (COD) levels were 55 and 320 mg/L in the influent, the TIN removal efficiencies were 89.7–92.4% in the step-feed mode and 48.1–59.5% in the conventional alternating A/O single-feed mode within a 360 min cycle. The pH and dissolved oxygen (DO) were used to optimize the process of denitrification and nitrification in the step-feed mode. The optimized operational condition was achieved by shortening the cycle time to 207 min, resulting in a nitrogen removal rate of 0.27 kg N/m^{3}d, which was much higher than those achieved using activated sludge systems. The dominant community in the aerobic granules was coccus-like bacteria, and filamentous bacteria were hardly found. Granules were well maintained throughout the 90 days of continuous step-feed operation.

1. Introduction

Nitrogen is one of the key nutrients causing eutrophication in water, and requires to be removed before discharge. Compared with the removal of phosphorus from wastewater, nitrogen can only be effectively and economically removed by biological methods. Biological nitrogen removal (BNR) generally involves aerobic nitrification with the conversion of NH_4–N to NO_2–N and/or NO_3–N by ammonia-oxidizing bacteria and/or nitrite-oxidizing bacteria, as well as denitrification with the conversion of NO_3–N and/or NO_2–N to molecular nitrogen by heterotrophic bacteria under the anoxic condition (Coelho et al., 2000). A sequence batch reactor (SBR) can carry out BNR in a single reactor based on the successive alternation of aerobic and anoxic reactions, which has attracted much interest in recent years for municipal and industrial wastewater treatment. However, during BNR, denitrification is normally the rate-limiting step because of the lack of sufficient organic carbon source to sustain a high denitrification rate, especially in treating ammonium-rich or low C/N wastewater (Chang and Hao, 1996; Chen et al., 2011; Shi et al., 2011). Consequently, a high concentration of NO_3–N or NO_2–N is often retained in the effluent, resulting in low nitrogen removal efficiency (Wang et al., 2009; Gao et al., 2011). To solve this problem, the step-feed SBR is proposed to improve nitrogen removal. The step-feed SBR can make good use of the influent COD as the carbon source required in the denitrification process (Wang et al., 2012), which means that the carbon source required to denitrify NO_3–N/NO_2–N formed in each aerobic phase is provided by the subsequent anoxic period influent. A step-feed SBR is reportedly both technologically and economically effective in enhancing nitrogen removal in activated sludge systems (Puig et al., 2004; Yang et al., 2007; Guo et al., 2008; Lemaire et al., 2009).

Typically, the activated sludge in the SBR is in the form of flocculent. Presently, a new form known as aerobic granular sludge exists and has been extensively studied over the past ten years (Beun et al., 1999; Qin and Liu, 2006; Adav et al., 2008; Muda et al., 2010; Verawaty et al., 2012). Compared with activated sludge, aerobic

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granules have a regular and compact physical structure, diverse microbial communities, good settling property, high biomass, and high resistance to organic loading shock. Thus, the aerobic granular sludge process is considered as one of the most promising processes for wastewater treatment. Generally, sufficient nitrifying biomass is difficult to obtain and maintain in a conventional activated sludge system, because nitrifying bacteria are sensitive to alterations in environmental factors and have low growth rates (Ochoa et al., 2002). However, the specific nitrification rate is higher in granular sludge than in flocculent. Aerobic granules also have a special layer of microbial structure that helps stimulate the growth of more denitrifying bacteria, eventually leading to better denitrification (Mosquera-Corral et al., 2005). Accordingly, many researchers have attempted to achieve a high nitrogen removal in the aerobic granule SBR system to treat synthetic wastewater (Kishida et al., 2006), and real municipal wastewater (Su et al., 2012). High nitrogen removal efficiencies ($\geqslant$90% and 81%) have been achieved under anoxic/oxic/anoxic and A/O conditions in the granular SBRs with the influent COD/N ratios of 10 and 4–8, respectively. Unfortunately, these results are not always the real events. Indeed, some experiments have obtained low nitrogen removal efficiencies (50–60%) in granular SBRs to treat the real municipal wastewater (Wang et al., 2009), and the synthetic wastewater (Yuan and Gao, 2010; Gao et al., 2011; Zhang et al., 2011). The main reason may be the deficient carbon source for denitrification. Thus, to obtain stable and high nitrogen removal efficiency, the application of the step-feed operational strategy and its nitrogen removal characteristics in an aerobic granular SBR system must be investigated.

To date, experimental research on granular SBR systems operated under the step-feed strategy is limited. Chen et al. (2011) studied the step-feed operation strategy in two granular SBRs using different particle sizes. They found that the nitrogen removal efficiency can reach 93% for 0.7 mm-sized granules and 95.9% for 1.5 mm-sized granules under 12 h of hydraulic retention time condition. Wang et al. (2012) explored the effects of step-feed on granulation and nitrogen removal performance of partial nitrifying granules in a two-phase step-feed SBR with influent COD and NH_4–N at 800 and 300 mg/L, respectively. The system achieved a nitrite accumulation rate of 93 ± 5% and total nitrogen (TN) removal efficiency of 70%. Nevertheless, the optimization of a granular SBR system with the step-feed strategy under continuous operation is still unreported. Thus, achieving high-rate nitrogen removal in a step-feed granular SBR under continuous operation requires further investigation. Accordingly, this study aimed to achieve high-rate nitrogen removal using the step-feed strategy in a granular sludge SBR system under continuous operation condition. Denitrification and nitrification durations were optimized using DO and pH as the control parameters under a four-phase step-feed condition. The stability of aerobic granules under continuous operation was determined as well.

2. Methods

2.1. Experimental SBR and operation

Experiments were carried out in a cylindrical column SBR (16 cm in diameter and 32 cm in high) with a working volume of 4.4 L and volumetric exchange ratio (VER) of 77%. The reactor was inoculated with activated sludge from the Gaobeidian municipal wastewater treatment plant in Beijing. The seeding sludge had a sludge age of 10 days, a mixed liquor suspended solids (MLSS) concentration of 4.1 g/L, and a sludge volume index (SVI) of 93 mL/g. Good and completely mature aerobic granules formed in 75 days. After the granulation was finished, nitrogen removal experiments were then conducted. A typical experiment was divided into three different stages. In stage I (1–28 d), the conditions were as follows: alternating A/O condition in the SBR with a 360 min cycle including 8 min feed, 60 min anoxic reaction, 260 min aerobic reaction, 3 min settling, 14 min decanting, and 15 min idling. In stage II (29–89 d), the conditions were as follows: four step-feeds with 30 min anoxic phase and 50 min aerobic phase in each feed for a 360 min cycle. In stage III (90–120 days), the conditions were as follows: optimization of nitrogen removal at four step-feeds and 207 min cycle. In stages II and III, the influent was introduced four times (2 min per feed) and sequentially fed into four anoxic phases with an equal volume of 0.85 L. The operational strategy of the three stages is shown in Fig. S1. The pH and DO were used to monitor the biological reaction process in the reactor; and they were automatically recorded online using the collecting data recorder at 1 min intervals. During aerobic reaction, air was introduced into the bottom of the reactor through a fine-bubble diffuser at a flow rate of 200 L/h. A slender agitator with a crescent-shaped vane was placed in the reactor, and the stirring speed was controlled at 120 rpm to avoid the granular sludge settling in the anoxic phase.

2.2. Synthetic wastewater

NH_4Cl and glucose were used as the nitrogen and carbon sources in the synthetic wastewater, respectively. The synthetic wastewater contained COD of approximately 320 mg/L, with the following typical composition: NH_4–N, 40 mg/L; PO_4^{3-}–P, 5 mg/L; NO_3–N, 15 mg/L (the NO_3–N in synthetic wastewater was mainly from groundwater); NaCl, 10 mg/L; and TIN, 55 mg/L. $NaHCO_3$ was used as buffering agent and potential inorganic carbon source for the nitrification process.

2.3. Analytical methods

COD, NH_4–N, NO_3–N, NO_2–N, MLSS, and SVI were tested according to the standard methods (APHA-AWWA-WEF, 2005). DO and pH were continuously monitored using Hach DO and pH meters. The particle size distribution was determined by a laser particle size analysis system (Malvern MasterSizer series 2000, Malvern instruments Ltd., Malvern, UK) within 0.02–2000 μm. The extracellular polymeric substances (EPS) content was qualified following the method described by Li and Yang (2007). The spatial structure of aerobic granules was observed by a scanning electron microscopy (SEM) system (FEIQUANTA 200).

3. Results

3.1. Overall performance of granular SBR

After complete granulation, the granular SBR system was operated for 120 days and the overall performance of the reactor operation is shown in Fig. 1. Stable and high COD and NH_4–N removal efficiencies were achieved at 92.4–95.2% and almost 100% in all three stages, respectively, regardless of changes in the operational strategy. This result demonstrated that the granular SBR technology can satisfy the strict effluent quality requirement for efficient COD and NH_4–N removal. However, high concentration of NO_3–N remained in the effluent, which resulted in low nitrogen removal of 48.1–59.5% in stage I. Subsequently, the TIN removal efficiency improved to 89.7–92.4% and 88.7–92.6% in stages II and III, respectively, after applying the step-feed operation mode. A quick and high nitrogen removal was observed on day 29, and TIN decreased to 5.8 mg/L in the effluent. No significant difference was observed between the nitrogen removal in stages II and III, although the