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A comparative study on denitrifying sludge granulation with different electron donors: Sulfide, thiosulfate and organics

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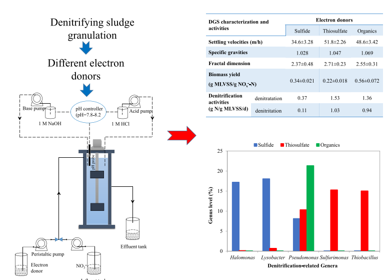
HIGHLIGHTS

- Denitrifying granular sludge (DGS) was cultivated with different electron donors.
- Longer time was required for sulfide-DGS cultivation.
- Characteristics among sulfide-, thiosulfate- and organics-DGS were compared.
- $S_2O_3^{2-}$-driven denitrifying sludge granulation is the most compact technology for BNR.

GRAPHICAL ABSTRACT



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ABSTRACT

A comparative study on denitrifying sludge granulation with different electron donors (sulfide, thiosulfate and organics) was carried out. Longer time was spent on sulfide-denitrifying granular sludge (DGS) cultivation (88 days) than thiosulfate- and organics-DGS cultivations (57 days). All the three DGS were characterized in terms of particle size distribution, sludge settling ability (indicated by sludge volume index and settling velocity), permeability (indicated by fractal dimension) and extracellular polymeric substances (EPS, including polysaccharide and protein) secretion. Sludge productions in the three DGS-reactors were also monitored. The key functional microorganisms in three granular reactors were revealed via high through-put pyrosequencing analysis. Batch tests were performed to measure the denitrification activities of each DGS, including both denitratation ($NO_3^- \rightarrow NO_2^-$) and denitritation ($NO_2^- \rightarrow N_2$). We found that thiosulfate-driven denitrifying sludge granulation (TDDSG) should be the most efficient and compact technology for effective BNR in municipal wastewater treatment. The findings of this study suggests the TDDSG could further increase the nitrogen removal potential in an enhanced sulfur cycle-driven bioprocess for co-treatment of wet flue gas desulfurization wastes with fresh sewage depending on three short-cut biological reactions, including: 1) short-cut biological sulfur

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reduction ($SO_4^{2-}/SO_3^{2-} \rightarrow S_2O_3^{2-}$); 2) thiosulfate-driven denitritation ($S_2O_3^{2-} + NO_2^- \rightarrow SO_4^{2-} + N_2\uparrow$); and 3) nitritation ($NH_4^+ + O_2 \rightarrow NO_2^-$).

1. Introduction

Biological nitrogen removal (BNR) process has been widely applied to remove nutrients (e.g. nitrogen) from wastewater in view of its effectiveness, environmental friendliness and energy conservation (Tchobanoglous et al., 2003). In BNR process, chemolithotrophic denitrification plays an essential role in the reduction of nitrate to nitrogen gas. With respect to denitrification, both heterotrophic denitrification (HD) and autotrophic denitrification (AD) have been reported extensively with each advantage. For example, higher nitrogen removal rate was demonstrated in HD reactor while lower sludge (bio-wastes) production was observed in AD reactor (Qian et al., 2015a; Oh et al., 2000). However, the floatation of the flocculent sludge and its washout from the conventional both HD and AD reactors are common problems during denitrification reactors' operation, which seriously deteriorates the nitrogen removal performance (Eiroa et al., 2005). Therefore, effective biomass retention in the denitrification reactor should be improved to maintain the stability of the sludge. And sludge granulation is one of the most promising technologies for the biomass retention (Val del Río et al., 2015).

Sludge granulation is an advanced environmental biotechnology in wastewater treatment based on the principle of biomass self-immobilization (Liu et al., 2003). Granular sludge (GS) appeared in the form of aggregates, is the collective of numerous cells. Various functional microorganisms stick to each other in the aggregates, which have the tightly compact structure and where diversified microbial communities are spatially distributed (Adav et al., 2008a,b). The characteristics of GS include abundant microbial biodiversity, high retainable biomass concentration, large specific density, low sludge yield, excellent sludge settling ability, and robust ability to withstand high pollutant loading rates (Liu et al., 2009). Therefore, granulation technology attracts tremendous academic interests in both industrial and domestic wastewater treatments. Despite the fact that denitrifying granular sludge (DGS) has been successfully cultivated in the lab-scale study (Val del Río et al., 2015), most of the these works focused on the effects of ratio of electron donors (e.g. organics and sulfide) to nitrogen (Yi et al., 2016), nitrogen loading rate (Li et al., 2013), nature of carbon sources (Lew et al., 2012), feeding strategy (Franco et al., 2006), hydraulic shear force (Xue et al., 2016), calcium/magnesium (Chen et al., 2015; Liu and Sun, 2011) on the DGS cultivation and stability. The comparison between the sludge granulations in HD and AD is missing in the literature.

As for AD, sulfide is the most commonly used electron donor and sulfide-based AD has been applied for simultaneous biodesulfurization and denitrification (Guo et al., 2016). Alternative to sulfide, thiosulfate is a potential electron donor for AD, characterized as high bioavailability (Cardoso et al., 2006). Recently, we proposed a sulfur cycle-based bioprocess for co-treatment of wet flue gas desulfurization (FGD) wastes with freshwater sewage. In this process, organics in the sewage are removed through biological sulfate/sulfite (via alkaline absorption of SO_2 from the fossil power plant) reduction. After the biological sulfate/sulfite reduction, three major compounds, i.e. sulfide, thiosulfate and organics residuals coexisted in the effluent. These compounds act as three types of electron donors in the subsequent denitrification reactor for BNR (Qian et al., 2015b) (the schematics of the co-treatment process of wet FGD wastes with freshwater sewage could be referred to Fig. S1). However, to the best of our knowledge, no detailed comparative studies on the denitrifying sludge granulation with these three electron donors, i.e. sulfide, thiosulfate and organics have been carried out so far.

In this study, three sequential batch reactors (SBR) were set up and operated in parallel with sulfide, thiosulfate and organics (i.e. acetate) as the electron donor for denitrification. The specific focuses are placed on: 1) the rate of sludge granulation fed with sulfide-, thiosulfate- and organics, respectively; 2) the nitrogen removal performance in the three DGS-SBR reactors and denitrification activities of the three DGS in the batch reactors 3) the physiochemical and biological characteristics of the three DGS.

2. Materials and methods

2.1. Experimental set-ups and DGS-SBR reactors' operation

Three lab-scale identical DGS-SBR reactors with effective volume of 2.2 L (height: 50 cm; diameter: 7.5 cm) were fabricated and set up, as shown in Fig. S2. The raw sludge was taken from a heterotrophic denitrification reactor of the modified A2O process in a municipal sewage treatment plant in Xi'an, China. About 1 L of raw sludge was seeded in each reactor, resulted in the initial biomass concentration of 1600 mg MLVSS/L (MLVSS: mixed liquor volatile suspended solids). 120 mg N/L sodium nitrate was used as the electron acceptor during the experiment. The whole reactors' operation period is divided into two phases. A 4-h operation cycle was employed in each reactor in Phase I (from Day 1–45), comprising 6 min feeding, 190-min reaction by mechanic mixing, 30-min settling, 10-min decanting and 4-min idling. The exchange ratio was 0.5 for three SBR reactors in each cycle, resulted in the hydraulic retention time (HRT) of 8 h. In Phase II (from Day 46–105), the cycle time was reduced to 2 h via decreasing the time of feeding, reaction, settling, decanting and idling to the half (see Table S1). Therefore, the HRT was dropped to 4 h and nitrogen loading rates were doubled in Phase II. The pH of each reactor was always controlled at 7.8 to 8.2 by alternatively adding 1 N HCl or 1 N NaOH solution whenever necessary (Fig. S2). As acetate was the main component of organic residues in the biological sulfate/suflite reducing reactor's effluent of the proposed sulfur cycle-driven bioprocess (Qian et al., 2015b), acetate was used as the organic source for HD sludge granular sludge cultivation in this lab-scale study. Concentrations of electron donor in each SBR reactor were 260 mg sulfide-S, 480 mg thiosulfate-S and 500 mg COD-acetate, which are all stoichiometrically sufficient to degrade 120 mg N/L of nitrate, as shown in Equations (1)–(3). The temperature of SBR reactors was kept at 25 ± 1 °C in an air-conditioned room. The detailed stock feeding composition (nutrients and trace metals) of the reactors is listed in Table S2. Samples from both influent and effluent of the three SBR reactors were taken regularly. Nitrate, nitrite, sulfide, thiosulfate, acetate, total suspended solids (TSS)/volatile suspended solids (VSS) concentrations were measured. Sludge samples from each reactor were also taken periodically for the analysis of mixed liquor suspended solids (MLSS)/mixed liquor volatile suspended solids (MLVSS),