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Enhanced elementary sulfur recovery with sequential sulfate-reducing, denitrifying sulfide-oxidizing processes in a cylindrical-type anaerobic baffled reactor

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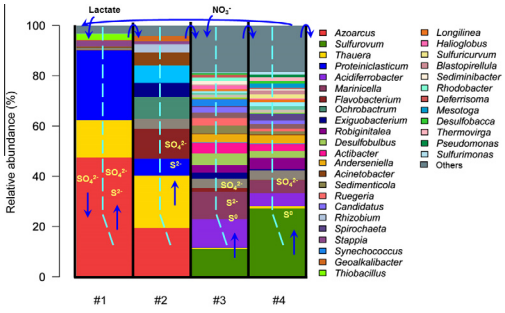
HIGHLIGHTS

- Simultaneous removals of SO_4^{2-}, NO_3^- and COD and recovery of elemental sulfur in ABR.
- Sulfate reduction and denitrifying sulfide removal were preceded sequentially.
- A high elemental sulfur recovery rate was obtained with SO_4^{2-}-S/NO_3^--N ratio of 5:5.
- Bacterial community analysis was conducted associated with SR and DSR processes.
- *DsrB* gene and *aprA* gene were abundant in SR and DSR units, respectively.

GRAPHICAL ABSTRACT



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ABSTRACT

Simultaneous removal of COD, SO_4^{2-} and NO_3^- and recovery of elemental sulfur (S^0) were evaluated in a four-compartment anaerobic baffled reactor (ABR) with separated functional units of sulfate reduction (SR) and denitrifying sulfide removal (DSR). Optimal SO_4^{2-}-S/NO_3^--N ratio was evaluated as 5:5, with a substantial improvement of S^0 recovery maintained at 79.1%, one of the highest level ever reported; meanwhile, removal rates of COD, SO_4^{2-} and NO_3^- were approached at 71.9%, 92.9% and 98.6%, respectively. Nitrate served as a key factor to control the shift of SR and DSR related populations, with the possible involvement of *Thauera* sp. during SR and *Sulfurovum* sp. or *Acidiferrobacter* sp. during DSR, respectively. *DsrB* and *aprA* genes were the most abundant during SR and DSR processes, respectively. Cylindrical-type ABR with the improved elemental sulfur recovery was recommended to deal with sulfate and nitrate-laden wastewater under the optimized SO_4^{2-}/NO_3^- ratio.

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1. Introduction

Sulfate and nitrate-laden wastewater generated from lot of industrial processes, such as pulp production, pharmacy, and petrochemical industry, constituted the serious threatens to human health and eco-systematic safety (Lens et al., 2003; Banu et al., 2008). Effective removal of sulfate and nitrate by application of microorganisms played crucial roles during wastewater treatment processes (Hao et al., 2014; Lu et al., 2014). Meanwhile, elemental sulfur, phosphorus and other nutrient recovery from wastewater are the hot research field during resource recovery processes (Raj et al., 2013; Yuan et al., 2014).

Conventional biological approach to completely remove the toxicity of sulfate-laden wastes consisted of two processes, sulfate reduction to sulfide by sulfate-reducing bacteria (SRB) and sulfide further oxidation to sulfur (S^0) by sulfide oxidation bacteria (SOB) (Wang et al., 2005). Recent studies found SOB could utilize nitrate as an electron acceptor during sulfide oxidation (denitrifying sulfide removal process, DSR) (Chen et al., 2008); studies on the simultaneous removal of organic carbon, sulfite and nitrate were conducted in either stirred tank reactors (CSTR) or expanded granular sludge bed (EGSB) reactor, which achieved a rather high recovery rate of S^0 (>90%) (Chen et al., 2009).

However, since SRB and SOB involved in diverse environmental niches, the integration of SRB and SOB reactors faced difficulty of the low S^0 conversion rate (Xu et al., 2012; Yuan et al., 2014). Xu et al. (2013) reported the intense bacterial competition during DSR process, which probably inhibited SOB and resulted in a low S^0 recovery rate. Therefore, proceeding of removal of organic carbon, sulfate and nitrate and simultaneous recovery of S^0 in integrated systems remained a technical challenge. Xu et al. (2012) attempted to introduce the limited quantity of DO into EGSB reactor, and found the limited quantity of DO stimulated activities of SOB but did not inhibit those of SRB, which achieved a rather high S^0 recovery rate of 71.8%. However, the precise control of DO level within this very narrow window (DO range of 0.10–0.12 mg L^{-1}) remains a technical difficulty requiring additional energy and cost.

Therefore, to obtain the high S^0 recovery rate from sulfate, separation of the sulfate reduction (SR) from DSR process is the key process to avoid the bacterial competition and guarantee the SOB activity. The anaerobic baffled reactor (ABR) with the separated reaction units connected in series possesses superiority in segregation of functional bacteria, compared with other anaerobic reactors (Uyanik et al., 2002; Zhu et al., 2008). Previously, some studies had reported the removal of sulfate and nitrate contained wastewater in ABR systems (Barber and Stuckey, 2000; Plumb et al., 2001), but up to now, there is no information on removals of organic carbon, nitrate and SO_4^{2-} for the S^0 recovery in ABR system.

The objective of this study was to demonstrate the feasibility of S^0 recovery during the sequential removal of organic matter, sulfate and nitrate in an ABR system with a separated functional unit of SR and DSR. The optimized SO_4^{2-}-S/NO_3^--N ratio was regulated to guarantee the high S^0 recovery rate. The molecular analysis on microbial community and functional genes was conducted to further investigate the interplays of functional populations and reaction mechanism. To the authors' best knowledge, this is the first study to describe the bioconversion of sulfate to S^0 coupled with the removal of organic matter and nitrate in ABR system.

2. Methods

2.1. Bioreactor configuration and operation conditions

Schematic of the applied ABR configuration were shown in Fig. 1. ABR system was cylindrical form (radius of 10 cm) and made of plexiglass, with four equal-volume discrete compartments and a total volume of 9.6 L. The four compartments were divided into two functional units, SR unit (compartment #1 and #2) and DSR unit (compartment #3 and #4). The flow started from influent of compartment #1 and sequentially passed through compartment #2 and #3 to #4 ultimately. In each compartment, regions of downcomer and upcomer were separated by a vertical baffle, with an angle of 45° at the bottom part to confirm the sufficient contact of wastewater and sludge. The volume ratio of upcomer and downcomer was 5:1. Peristaltic pumps were used to control the influent and effluent ratio, with a reflux ratio of 5:1. ABR was operated with a fixed HRT of 24 h. Compared with other ABR systems, the cylindrical-type ABR can largely reduce the high load in the first compartment and maintain the fine microbial activity. The above designed ABR has been authorized as a Chinese patent (201310484595.8).

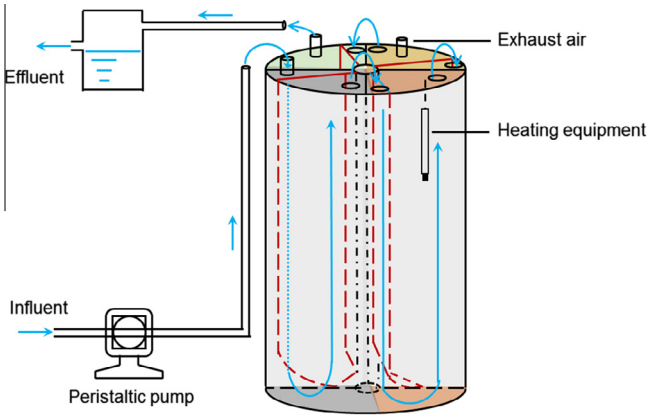

Fig. 1. Schematic diagram of the applied pilot-scale anaerobic baffled reactor (ABR).

The applied seed sludge was collected from the anaerobic sludge thickener at the WenChang Wastewater Treatment Plant, Harbin, China. The influent concentration (COD/SO_4^{2-}-S/NO_3^--N) at three stages was shown in Table 1. The influent (L^{-1}) in compartment #1 contained: SO_4^{2-} (500 mg), sodium lactate (with the COD of 1000 mg), Ca^{2+} (25 mg), Mg^{2+} (10 mg) and trace element. Bicarbonate (1–2 g L^{-1}) was employed to maintain the influent pH of 8.0 ± 0.3. The trace element solution was fed into the influent with the detailed composition described by Chen et al. (2009). Nitrate was applied to compartment #3, with the concentration of 132.2 mg L^{-1}, 326.1 mg L^{-1} and 504.4 mg L^{-1}, at stage I, II and III, respectively. The resulted S/N ratios at three stages were 5:2, 5:5 and 5:8, respectively (Table 1).

2.2. Analytical methods

Influent and effluent samples (3–10 mL) were collected from inlet and outlet of the reactor and stored in −4 °C refrigerator before went for chemical analysis. Samples (3–10 mL) from the middle of the four compartments at steady running state were harvested with a sterilized sample spoon and stored in a 50 mL sterile plastic test tubes at −80 °C before DNA and RNA extraction.

COD was measured according to US standard methods of water and wastewater measurement (APHA, 1998). Sulfide concentration (including H_2S, HS^{-1} and S^{2-}) was determined according to the methylene blue method (Trüper and Schlegel, 1964). Concentrations of SO_4^{2-}, $S_2O_3^{2-}$, SO_3^{2-}, NO_3^- and NO_2^- were measured by an ion chromatography (ICS-90A, Dionex, USA) after filtrated with 0.45 μm of the millipore filter. A pH/ORP meter (No. FE20, Merrler Toledo, China) was used to determine the pH and oxidation–reduction potential (ORP) of liquid samples. Production of