

## Study of a combined heterotrophic and sulfur autotrophic denitrification technology for removal of nitrate in water

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### ABSTRACT

A combined two-step process of heterotrophic denitrification in a fluidized reactor and sulfur autotrophic denitrification processes (CHSAD) was developed for the removal of nitrate in drinking water. In this process, the advantage of high efficiency of heterotrophic denitrification with non-excessive methanol and the advantage of non-pollution of sulfur autotrophic denitrification were integrated in this CHSAD process. And, this CHSAD process had the capacity of pH balance and could control the concentration of  $\text{SO}_4^{2-}$  in effluent by adjusting the operation condition. When the influent nitrate was  $30 \text{ mg NO}_3^- \text{--N/L}$ , the reactor could be operated efficiently at the hydraulic retention time (HRT) ranging from 20 to 40 min with C:N ratio ( $\text{mg CH}_3\text{OH}:\text{mg NO}_3^- \text{--N}$ ) of 2.0 (methanol as carbon source). The nitrate removal was nearly 100% and there was no accumulated nitrite or residual methanol in the effluent. The effluent pH was about 7.5 and the sulfate concentration was lower than  $130 \text{ mg/L}$ . The maximum volume-loading rate of the reactor was  $2.16 \text{ kg NO}_3^- \text{--N}/(\text{m}^3 \text{ d})$ . The biomass and scanning electron microscopy graphs of biofilm were also analyzed.

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### 1. Introduction

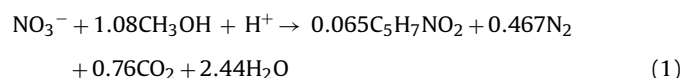
Nitrate is a wide spread contaminant of ground and surface waters due to excessive use of nitrogenous fertilizer in agricultural industry and inappropriate disposal of untreated sanitary and industrial wastes [1]. Nitrate can cause methemoglobinemia when ingested by infants, and could cause carcinoma, malformation and mutation when transformed into nitrosoamines [2–4]. The ground water from 10 to 25% of the water-supply wells in large region of the US exceeds the maximum contaminant level (MCL) for nitrate ( $10 \text{ mg NO}_3^- \text{--N/L}$ ) [5]. In China, the pollution of nitrate in ground water is more severe. The nitrate concentration of groundwater in some rural areas exceeds  $130 \text{ mg NO}_3^- \text{--N/L}$ . Ground water is a universal, and in some cases exclusive, drinking-water source used by both humans and livestock in rural and suburban areas. Therefore, the remediation of nitrate-contaminated groundwater is one of the targets urgently confronted.

Current chemical technologies for removal of nitrate like ion exchange and reverse osmosis are not selective to nitrate, generate secondary brine wastes and require generation of used medium [6]. Biological denitrification technology, which comprises heterotrophic denitrification [7–11] or autotrophic denitrification

[12–19], is one of the most efficient methods for nitrates removal from water. The heterotrophic denitrification system uses organic compounds, such as methanol [7–9] and ethanol [10,11], as carbon source. Its main advantages are high denitrifying rate and treatment capacity. However, nitrite will be produced and accumulated in water when the added organic is stoichiometrically insufficient [9]. Contrarily, the residual organic compound will pollute the treated water when the added organic is excessive [10].

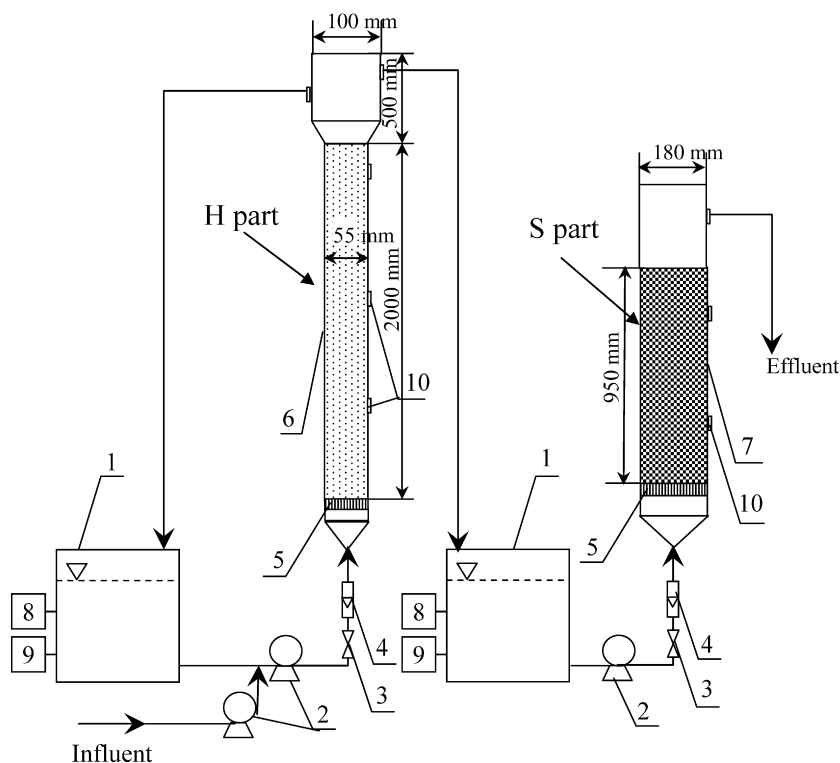
As for autotrophic denitrification, both elemental sulfur [12–17] and hydrogen gas [18,19] can be used as ideal electron donor. Sulfur limestone autotrophic denitrification (SLAD) systems have been studied widely in Europe and USA. In such process, elemental sulfur is used as electron donor while limestone is used to adjust the pH.  $7.54 \text{ mg/L}$  sulfate will be produced when  $1 \text{ mg NO}_3^- \text{--N/L}$  is removed. There will be an increase in hardness in the treated water because  $\text{Ca}^{2+}$  is added due to the pH adjustment by limestone [20].

In order to take full advantage of heterotrophic denitrification and sulfur autotrophic denitrification, a new combined two-step process of heterotrophic and sulfur autotrophic denitrification processes (CHSAD) was developed. In the CHSAD reactor, nitrate is firstly reduced in H part, and the residual subsequently is reduced in S part. In H part, the nitrate is denitrified by heterotrophic denitrification when methanol is dosed as organic carbon sources.  $\text{H}^+$  was consumed. The reaction is shown in Eq. (1) [21]:



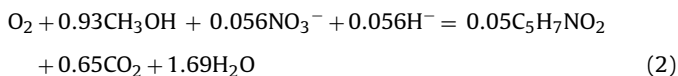
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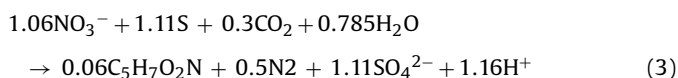
**Fig. 1.** Schematic of the CHSAD reactors: (1) recycle reservoir, (2) pump, (3) cutoff valve, (4) liquid rotameter, (5) distributor, (6) fluidized bed of heterotrophic denitrification, (7) bed of sulfur autotrophic denitrification, (8) temperature control system, (9) pH analyzer, and (10) sampling sites.

The stoichiometric ratio of C:N (mg CH<sub>3</sub>OH:mg NO<sub>3</sub><sup>-</sup>-N) for complete denitrification using methanol as organic carbon source was 2.47 according to the reaction. In the practical process, the C:N ratio will be higher than 2.47 due to the presence of oxygen in the water. The bacteria will consume some methanol as shown in reaction (2)



In this study, the feed DO was normally below 0.5 mg/L, and was controlled below 1 mg/L because the nitrogen gas was purged in the experiments.

The advantage of heterotrophic denitrification is its higher efficiency, while the disadvantage is the secondary pollution from the residual methanol. In addition, the pH will increase according to Eq. (1). Denitrification technologies will have to be developed so that the ratio of CH<sub>3</sub>OH to NO<sub>3</sub><sup>-</sup>-N is below 2.47 for the complete removal of the residual nitrate. In this study, we combined the heterotrophic denitrification with sulfur autotrophic denitrification. In S part, the autotrophic denitrification took place as shown in Eq. (3). In this process, SO<sub>4</sub><sup>2-</sup> and H<sup>+</sup> will be produced [17].



It can be seen from Eq. (1) that 1.0 mol H<sup>+</sup> would be consumed in H part when 1 mol NO<sub>3</sub><sup>-</sup>-N was denitrified, and as a result the pH in effluent will decrease. On the other hand, 1.09 mol H<sup>+</sup> will be provided when 1 mol NO<sub>3</sub><sup>-</sup>-N is reduced to nitrogen gas according to Eq. (3). Thus the pH can be better maintained. The combined technology could achieve high nitrate removal efficiency, no residual methanol and no nitrite accumulation. In addition, the concentration of SO<sub>4</sub><sup>2-</sup> will be controlled by adjusting the nitrate load proportion of H part and S part.

Only very few studies [22–24] focus on combined heterotrophic and autotrophic process for waste water denitrification. For drinking water, there was no report on combined heterotrophic and autotrophic denitrification process. Therefore, the main objectives of this research are (1) to develop the CHSAD process; (2) to verify the feasibility of this process; and (3) to present the applicable operation conditions of this process for nitrate removal from water.

## 2. Materials and methods

### 2.1. Experimental set-up

Experiments were performed in the apparatus shown in Fig. 1. The reactor consisted of H part and S part, and the total effective liquid volume of the reactor was 19.6 L.

The fluidized bed in H part constructed from the plexiglas had an i.d. of 55 mm and a height of 2000 mm. It was ended by a disengaging cap with an i.d. of 100 mm and a height of 500 mm. Anthracite (Gongyi, China) with a diameter of 1.0–1.2 mm was used as carriers for the microorganisms. The effective volume of H part was 4 L.

S part included a cylindrical plexiglas with an i.d. of 180 mm and a height of 2400 mm; its effective volume was 15.6 L. Sulfur granules obtained from Luoyang Sunrise Industrial of China had a diameter of 5–7 mm. The height of the sulfur granules packed bed was 950 mm. The liquid flow rate measured with a rotameter was controlled by a cutoff valve. The temperature was controlled by an electric heater coupled with a contact thermometer.

The synthetic water with 30 mg NO<sub>3</sub><sup>-</sup>-N/L as an influent in our study was prepared by the water from the 9th Beijing Water Treatment Plant and certain amount of NaNO<sub>3</sub>. The water had IC (inorganic carbon) of 8 mg C/L, SO<sub>4</sub><sup>2-</sup> concentration of about 40 mg/L and pH of 7.6. All chemicals used in this experiment were analytical grade (Beijing Agent Plant, Beijing, China). The influent was first pumped from feed tank to H part and then to S part; the flow rate ranged from 2 to 12 L/h. The feed was purged with

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