



Simultaneous removal of perchlorate and nitrate in a combined reactor of sulfur autotrophy and electrochemical hydrogen autotrophy



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HIGHLIGHTS

- S^0 - and H_2 -autotrophy for NO_3^- and ClO_4^- reduction were investigated in a reactor.
- Autotrophic denitrifying process favored over autotrophic perchlorate reduction.
- H^+ from S^0 autotrophy process was consumed by electrochemical H_2 autotrophy.
- Some autotrophic denitrifier can use both NO_3^- and ClO_4^- as electron acceptor.
- Microbial communities at different positions in the reactor were analyzed.

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ABSTRACT

The removal of perchlorate (ClO_4^-) and nitrate (NO_3^-) from drinking water was investigated in a combined reactor of sulfur autotrophy (S-compartment) and electrochemical hydrogen autotrophy (H-compartment). The removal efficiencies of NO_3^- and ClO_4^- in the S-compartment and H-compartment were affected by hydraulic retention time (HRT) and current intensity, respectively. The sulfur- and hydrogen-autotrophic denitrifying process favored over the process of sulfur- and hydrogen-autotrophic perchlorate reduction in the combined reactor. The longer HRT could lead to sulfur (S^0) disproportionation due to the increase of the contact time between water and S^0 particle. The H^+ generated from S-compartment could be reduced as H_2 by electrochemical process in H-compartment, and the generated H_2 as an electron donor was utilized to reduce NO_3^- and ClO_4^- by hydrogen autotrophic reduction. The oxidation reduction potential (ORP) in the effluent from S-compartment and H-compartment were below -180 mV, suggesting good anaerobic conditions for the reduction of NO_3^- and ClO_4^- in the combined reactor. Some sulfur- and hydrogen-autotrophic denitrifying bacteria could use both NO_3^- and ClO_4^- as electron acceptors in the combined reactor. The DGGR profile illustrated that some variations were found in the microbial community at different locations of the combined reactor.

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1. Introduction

Perchlorate (ClO_4^-) has been widely used in solid rocket fuels, missiles, explosives, pyrotechnics, and fireworks [1–3]. ClO_4^- contamination in surface water and groundwater has become a significant environmental concern in recent years. ClO_4^- can disrupt thyroid function by interfering with body's intake of iodine, which inhibits the production of thyroid hormone [4]. Nitrate (NO_3^-) is

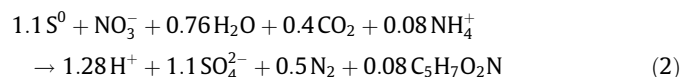
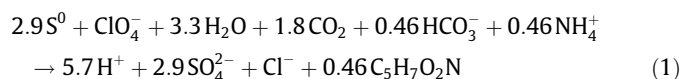
often a co-contaminant with ClO_4^- in surface water and groundwater, which mainly comes from agricultural runoff and wastewater discharge. The consumption of water contaminated by NO_3^- can cause methemoglobinemia in infants, malformation and mutation [5].

The current technologies for ClO_4^- and NO_3^- removal from drinking water and groundwater include ion exchange, photocatalytic reduction, membrane technology, absorption and biological reduction [6–13]. Ion exchange and biological reduction are regarded as the most cost-effective technologies for the treatment of ClO_4^- and NO_3^- contaminated water. The major disadvantage of ion exchange is that it can generate ClO_4^- - and NO_3^- -laden brine, which requires specialized disposal and/or treatment. Biological reduction may

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completely transform ClO_4^- and NO_3^- into innocuous Cl^- and N_2 , respectively. Some biological processes using different organic substrates as electron donors have been proved to be highly effective for ClO_4^- and NO_3^- removal [14–17]. However, the residual organic substrate in the effluent can stimulate microbial growth in water distribution system and contribute to the formation of potentially toxic trihalomethanes during disinfection by chlorination [18]. To settle the above-mentioned problems, autotrophic reduction of ClO_4^- and NO_3^- has been carried out by using inorganic matters as electron donors [19–21]. Hydrogen (H_2) autotrophic reduction of ClO_4^- and NO_3^- has been evaluated in some continuous flow bioreactors [19,22]. Although H_2 is a good energy substrate for ClO_4^- and NO_3^- reduction, it is difficult to handle in bulk quantities and is publicly perceived to represent a significant disaster threat due to its inherent explosive nature. Granular sulfur (S^0) in packed bed bioreactors has been successfully used as an electron donor in the autotrophic reduction of ClO_4^- and NO_3^- [20,23–26]. As granular S^0 is insoluble in water, it can provide a slow-release supply of electron on demand and offer low expense cost as well as low maintenance cost. The stoichiometric equations for the S^0 autotrophic reduction of ClO_4^- and NO_3^- are respectively shown as follows [24,26]:



The main disadvantage for sulfur autotrophic ClO_4^- and NO_3^- reduction is the formation of sulfate (SO_4^{2-}) and H^+ in the effluent. Limestone is often used as a low-cost alkalinity source, whereas it can increase the hardness of the effluent due to the release of Ca^{2+} during limestone dissolution. To make up for the deficiency of sulfur autotrophic ClO_4^- and NO_3^- reduction, a combined reactor of sulfur autotrophy and electrochemical hydrogen autotrophy was used to remove ClO_4^- and NO_3^- in the present study. In the combined reactor, the H^+ generated from sulfur-autotrophy ClO_4^- and NO_3^- reduction could be consumed by the bioelectrochemical hydrogen-autotrophy ClO_4^- and NO_3^- reduction to achieve neutralization, and the SO_4^{2-} concentration in the effluent can be controlled by adjusting the ClO_4^- and NO_3^- loading of sulfur autotrophy and electrochemical hydrogen autotrophy.

The main objectives of the present study were (1) to evaluate the feasibility of ClO_4^- reduction by sulfur- and hydrogen-autotrophic denitrifying bacteria; (2) to investigate the effect of hydraulic retention time (HRT) and current intensity on ClO_4^- and NO_3^- reduction in a combined reactor of sulfur autotrophy and electrochemical hydrogen autotrophy; (3) to analyze the variation of microbial community along the flow path in the combined reactor.

2. Materials and methods

2.1. Experimental set-up

The schematic diagram of an experimental set-up is shown in Fig. 1, which consists of sulfur autotrophy (S-compartment) and electrochemical hydrogen autotrophy (H-compartment). The S-compartment was a cylindrical plexiglass column with 7.5 cm in diameter and 40 cm in height, which was filled with S^0 granules of 3.0–4.0 mm in diameter. A cylindrical stainless column with 7.5 cm in diameter and 25 cm in height was used as the cathode in the H-compartment. A graphite rod with 2.5 cm in diameter and 25 cm in height was used as the anode in the H-compartment, which was located in the center of the cylindrical

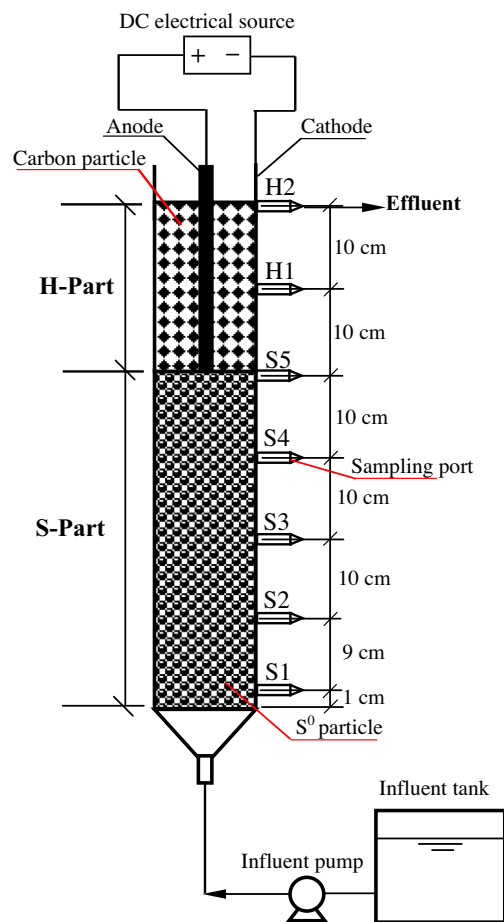


Fig. 1. Schematic diagram of an experimental set-up.

stainless column. The space between the cathode and the anode in the H-compartment was filled with carbon particle with 3.0–4.0 mm in diameter. Before the synthetic wastewater was fed to the combined reactor, it was firstly deoxidized by injecting nitrogen gas in the influent tank. A peristaltic pump was used to feed the influent into the combined reactor, and direct current was supplied to the H-compartment by a silicon rectifier. The Chemical processes of NO_3^- and ClO_4^- reduction in the S-compartment and H-compartment is shown in Fig. 2. The water sampling ports were mounted at the height of 1 cm (S1), 10 cm (S2), 20 cm (S3), 30 cm (S4), 40 cm (S5), 50 cm (H1) and 60 cm (H2) from the bottom of the combined reactor. In addition, the S^0 granules in the S-compartment were not replenished during the whole operational periods.

2.2. Inoculated sludge and synthetic wastewater

The inoculated sludge was taken from an anaerobic tank of Lichunhe wastewater treatment plant in Qingdao city, China. The synthetic wastewater fed to the combined reactor was prepared daily by adding NaNO_3 , $\text{NaClO}_4 \cdot \text{H}_2\text{O}$, and nutrient medium to the tap water. The compositions of nutrient medium were shown as follows: NH_4HCO_3 0.410 g/L, NaHCO_3 2.700 g/L, $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ 0.100 g/L, $\text{Ca}(\text{OH})_2$ 0.005 g/L, and trace element solution 1 mL/L. The trace element solution consisted of the following components (g/L): $\text{Na}_2\text{SeO}_3 \cdot 5\text{H}_2\text{O}$ 0.1, H_3BO_3 0.05, $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ 0.415, $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ 0.113, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ 2.8, $\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$ 0.5, $\text{CoSO}_4 \cdot 12\text{H}_2\text{O}$ 2.36, $\text{KAl}(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$ 0.175, EDTA 1.0, $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ 0.106, $(\text{NH}_4)_6\text{Mo}_7\text{O}_{24} \cdot \text{H}_2\text{O}$ 0.05, and $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ 0.157.

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