



Complete bromate and nitrate reduction using hydrogen as the sole electron donor in a rotating biofilm-electrode reactor



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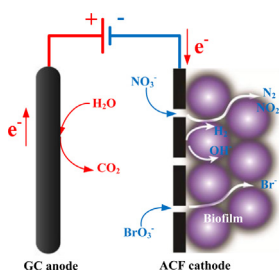
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HIGHLIGHTS

- Cathode of RBER was designed to automatically rotate.
- Simultaneous bromate and nitrate removal was achieved by auto-hydrogenotrophic reduction.
- The maximum bromate reduction rate estimated by the Monod equation was 109.12 $\mu\text{g/L h}$.
- An electron transfer process and main reaction mechanism in RBER was explored.

GRAPHICAL ABSTRACT

Main mechanism of simultaneous bromate and nitrate removal in the RBER.



ARTICLE INFO

Article history:

Received 8 July 2015

Received in revised form 1 December 2015

Accepted 26 December 2015

Available online 31 December 2015

Keywords:

Bromate

Nitrate

Hydrogen

Electron donor

Rotating biofilm-electrode reactor

ABSTRACT

Simultaneous reduction of bromate and nitrate was investigated using a rotating biofilm-electrode reactor (RBER) with graphite carbon (GC) rods as anode and activated carbon fiber (ACF) bonded with steel ring as cathode. In RBER, the community of denitrifying bacteria immobilized on the cathode surface could completely utilize hydrogen (H_2) as the electron donor, which was internally produced by the electrolysis of water. The short-term test confirmed that the RBER system could reduce 150–800 $\mu\text{g/L}$ bromate to below 10 $\mu\text{g/L}$ under autotrophic conditions. The reduced bromate was considered to be roughly equivalent to the amount of bromide in effluent, indicating that bromate was completely reduced to bromide without accumulation of by-products. The long-term test (over 120 days) showed that the removal fluxes of bromate and nitrate could be improved by increasing the electric current and decreasing the hydraulic retention time (HRT). But nitrite in effluent was significantly accumulated when the electric current was beyond 10 mA and the HRT was less than 6 h. The maximum bromate reduction rate estimated by the Monod equation was 109.12 $\mu\text{g/L h}$ when the electric current was 10 mA and HRT was 12 h. It was proposed that the electron transfer process in RBER produced H_2 on the surface of the ACF cathode, and the microbial cultures attached closely on the cathode which could completely utilize H_2 as electron donors for reduction of bromate and nitrate.

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1. Introduction

Bromate (BrO_3^-) is a disinfection by-product from the ozonation or advanced oxidation of water containing bromide (Br^-). It has been experimentally demonstrated that KBrO_3 induces renal

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cell tumors, peritoneal mesothelioma, and follicular cell tumors of the thyroid [1,2]. Hence, the International Agency for Research on Cancer (IARC) and the World Health Organization (WHO) has classified bromate as a group B-2 carcinogen (a suspected human carcinogen). Currently, the European Union and the U. S. Environmental Protection Agency (EPA) have set a maximum contaminant level of 10 $\mu\text{g/L}$ (0.078 $\mu\text{mol/L}$) for bromate in drinking water [3]. Therefore, it is necessary to control or remove bromate from bromate-contaminated water. In the past decades, a lot of methods have been successfully implemented to remove bromate, such as adsorption process [4,5], membrane separation [6,7], catalytic decomposition [8–10] and biological reduction [11,12]. Among them, biological reduction is considered to be the a promising alternative due to its ability of reducing bromate to relatively innocuous bromide in a cost-effective way [13].

Previous research showed that bromate could be reduced to bromide by denitrifying bacteria like *Pseudomonas* spp. possibly via co-metabolic action of nitrate reductase [14]. The biological denitrification, either autotrophic or heterotrophic, has been proposed for nitrate and/or bromate removal [15,16]. The heterotrophic denitrification requires the organic carbon sources, such as ethanol, glucose and acetate, as electron donor for the respiration and growth of microorganisms. These organic substrates could potentially leave organic residuals which are easy to form the toxic disinfection byproducts during chlorination [17]. By contrast, autotrophic denitrification does not require organic supplementation, in which some inorganic compounds act as electron donor instead of organic substrates. The elemental sulfur (S°) and sulfide have been reported to be used as the electron donor for the reduction of bromate and/or nitrate in sulfur-based autotrophic denitrification due to its low cost and availability [18,19], but it need some extra steps to remove the reaction residues. Comparing to the other electron donors, the hydrogen gas (H_2) as an attractive alternative electron donor possess the characteristic of harmless and naturally cleaning, and hence, it has been widely applied in the microbial reduction of oxidized inorganic compounds, including perchlorate [20,21], nitrate [22,23], sulfate [24] and their mixture [25–27].

Although H_2 has been proven to be an effective electron donor, the present application in practice is subject to some limit owing to its low solubility in water (1.6 mg/L at 20 °C) and explosion characteristic. To overcome these shortcomings, Nerenberg et al. designed a new reactor, the hollow-fiber membrane biofilm reactor (MBfR), to safely and efficiently deliver H_2 to the biofilm for reduction of oxidized contaminants in drinking water [28]. In the MBfR, an autotrophic biofilm naturally grows on the outside wall of the membrane fibers, and the H_2 electron donor diffuses through the wall of membrane fibers to meet the contaminant electron acceptor at the interface. Because of the counter-current transport of H_2 and the oxidized contaminant in the biofilm, H_2 -utilization efficiency could achieve nearly 100% [29]. However, H_2 has to be supplied externally in MBfR and the risk of hydrogen storage still exists. In addition, some reports showed that the internal (electrochemical or chemical) production of H_2 could be more effectively utilized by bacteria than that of external supplied [30]. To overcome the above-mentioned disadvantages, biofilm-electrode reactor (BER) combining biological and electrochemical method has developed and employed to remove nitrate based on the hydrogen autotrophic denitrification. In BER, H_2 is generated on site by electrolysis of water and completely utilized as the electron donor by the denitrifying bacteria immobilized on the cathode surface, avoiding the waste of excessive H_2 in the case of external addition [31]. It has been demonstrated that BER is a promising method for nitrate removal from drinking water [30,32,33], but none of studies have been dedicated to the simultaneous removal of bromate and nitrate by BER.

In this research, an auto-hydrogenotrophic rotating biofilm-electrode reactor (RBER) was developed to remove bromate and nitrate simultaneously by autotrophic denitrification using H_2 as the sole electron donor. The cathode of RBER was designed to automatically rotate so as to enhance the immobilization of biofilm on the cathode and achieve the complete mixing condition. The main objectives of this research were to (1) determine whether the autotrophic denitrifying bacteria in RBER could reduce bromate to below the emission standard of 10- $\mu\text{g/L}$ treatment objective; (2) investigate the effect of electric current and HRT on bromate and nitrate reduction; and (3) explore the electron transfer process and the mechanism of simultaneous reduction of bromate and nitrate in the RBER.

2. Materials and methods

2.1. Experimental set-up

The schematic view of rotating biofilm-electrode reactor (RBER) is shown in Fig. 1. The RBER consisted of polyethylene plastic cylinder (12 cm in diameter and 14 cm in height) as an electrolytic tank, an activated carbon fiber (ACF) bonded with steel ring as cathode (8 × 8 cm) and two graphite carbon (GC) rods as anode ($\phi 5 \times 150$ mm). The cathode was designed to automatically rotate through the motor stirrer and the rotating rate is constant at 14 ± 1 r/min, which enhances the immobilization of biofilm on the cathode and achieve the complete mixing condition. The reactor was tightly sealed to maintain anaerobic condition with the effective working volume of 1.21 L. Electric current or electrode potential was applied by using a DC power supply (Rek PS-303D, China) with a voltage range of 0–30 V. The pH of influent solution was adjusted to 7.2 ± 0.2 with either 0.5-M NaOH or 0.5-M H_2SO_4 .

2.2. Synthetic medium

The medium (per liter of ultrapure water) contained following mineral salts (analytical grade): 1.55 g/L K_2HPO_4 , 0.85 g/L KH_2PO_4 , 0.10 g/L $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, 0.25 g/L NaHCO_3 . The aliquots of trace mineral containing metal-chelator has been described previously [34]. NaNO_3 , NaBrO_3 and the synthetic medium were added in tap water as synthetic water to achieve the concentrations which are given in Table 1. The medium was maintained at 35 ± 2 °C via water bath during all periods. Before feeding, nitrogen (N_2) gas was continuously sprayed to make sure the solution under low DO condition (0.4 ± 0.1 mg/L).

The feed solution is deoxygenated passing through N_2 gas only at the early period (0–15 days) in order to accelerate the anaerobic bacteria growth and bifilm formation. After the acclimation accomplished, the synthetic medium flowed into the RBER directly without deoxygenating by N_2 gas.

2.3. Inoculum and acclimation

200 mL anaerobic sludge was taken from the second municipal wastewater treatment plant (Changsha, China) which was used as the inoculums. The inoculums were acclimated in RBER for 5 days in the batch mode, feeding the above-mentioned medium every day. Then, the microbe was further acclimated in continuous mode, feeding synthetic water containing 25 mg/L nitrate. The hydraulic retention time (HRT) and an applied electric current was set at 12 h and 10 mA, respectively. About 15 days later, the nitrate removal efficiency was higher than 75% and a dark grey biofilm covered the activated carbon fibers, indicating that the acclimation accomplished.

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