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Single microbial fuel cell reactor for coking wastewater treatment: Simultaneous carbon and nitrogen removal with zero alkaline consumption

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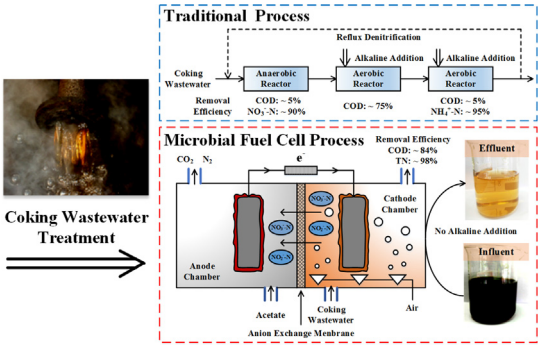
HIGHLIGHTS

- A single microbial fuel cell reactor was used for coking wastewater treatment.
- Simultaneous carbon and nitrogen removal is demonstrated.
- There is no need for addition of alkali reagents during the treatment.
- The current is critical in affecting the degradation performance of the reactor.
- Heterotrophs, nitrifiers, and denitrifiers are enriched in the reactor.

GRAPHICAL ABSTRACT



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ABSTRACT

The use of several individual reactors for sequential removal of organic compounds and nitrogen, in addition to the required alkaline addition in aerobic reactors, remain outstanding technical challenges to the traditional biological treatment of coking wastewater. Here, we report the utilization of a single microbial fuel cell (MFC) reactor that performs simultaneous carbon and nitrogen removal with zero alkaline consumption, as evidenced by the results of the batch-fed and continuous-flow experiments. The MFC exhibited faster reaction kinetics for COD and total nitrogen (TN) removal than the same configured reactor analogous to the traditional aerobic biological reactor (ABR). At a hydraulic retention time (HRT) of 125 h, the efficiencies of COD and TN removal in the MFC reached 83.8 ± 3.6% and 97.9 ± 2.1%, respectively, much higher than the values of 73.8 ± 2.9% and 50.2 ± 5.0% obtained in the ABR. Furthermore, the degradation in the MFC of the main organic components, including phenolic compounds (such as phenol, 2-methylphenol, 3-methylphenol, 4-methylphenol, and 2,4-dimethlyphenol) and nitrogenous heterocyclic compounds (such as quinolone, pyridine, indole, and isoquinolone) was greater than that in the ABR. The enhancing effect was attributed to the ability of the MFC to self-adjust the pH. It was also manifested by the increased abundances of heterotrophs, nitrifiers, and denitrifiers in the MFC. The correlations between the current density and the rates of COD and TN removal suggest that the extent of the current from the anode to the cathode is a critical parameter for the overall performance of MFCs in the treatment of coking wastewater.

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1. Introduction

Coking wastewater is usually characterized by complicated composition, high chemical oxygen demand (COD), elevated concentrations of ammonium, and severe toxicity (Wei et al., 2012; Huang et al., 2016; Joshi et al., 2016). Treating the huge amounts of coking wastewater generated by coke production (approximately 2.85×10^8 m^3 per year) is a great challenge in China, which is the largest coke producer in the world (almost 470 million tons of coke per year). Our survey of 80 currently operating coking wastewater treatment plants (CWTP) in China show that the main biological treatments for the removal of organic carbon and total nitrogen (TN) are based on anaerobic-anoxic-oxic (A/A/O), anaerobic-oxic-anoxic-oxic (A/O/A/O), or oxic/anoxic/oxic (O/A/O) processes. A careful examination of the biological systems of these plants, however, reveals that there are at least two technical bottlenecks adding to the investment and operational costs of treatment. The first one lies in the construction of a series of biological reactors that sequentially remove organic compounds and nitrogen. For example, the CWTP of Shaogang (Guangdong Province of China), designed by our group, has an average treatment capacity of 2000 m^3 per day; the plant integrates three tanks, the first aerobic tank for the removal of phenols and other compounds, the second aerobic tank for nitrification, and the third anaerobic tank for denitrification (Zhang et al., 2012). The second one requires considerable consumption of alkali reagents, which are used to neutralize the excess protons produced during nitrification and other aerobic oxidation processes. A similar example from Shaogang CWTP indicates that the addition of at least 0.58 kg NaOH/m^3 d to the aerobic tank is needed to adjust the pH.

Due to the growing attention to sustainable, versatile, and efficient biological systems for the treatment of industrial wastewater laden with high-concentration COD and ammonium, the interest in developing new technologies that overcome the aforementioned bottlenecks is significant. From an engineering point of view, the outstanding challenge is to achieve simultaneous nitrification and denitrification (SND) in a single aerobic reactor. Two SND concepts have received considerable attention. One of them relies on the gradient descent of the oxygen concentration from the outer to the internal areas in the granular sludge or biofilm-based reactor, creating a separate aerobic zone for nitrification and an anoxic zone for denitrification (Di Bella and Torregrossa, 2013; Corsino et al., 2016). The other concept is related to the discovery of certain groups of bacteria that are capable of heterotrophic nitrification and aerobic denitrification and are thus able to aerobically convert ammonium to nitrogen (Gokce et al., 1989). Despite the benefits, the enrichment of denitrifiers under aerobic conditions remains difficult, which may limit their further application in wastewater treatment. We previously reported a promising alternative that can technologically enable SND by using a double-chamber anion microbial fuel cell (MFC) with an anion-exchange membrane (Feng et al., 2015). The principle of action lies in the fact that the nitrate produced as a result of aerobic nitrification can quickly move from the cathode chamber to the anode chamber, where anaerobic denitrification with organic substrates occurs. The rates of nitrate diffusion and conversion to nitrogen in the anode chamber are much faster than the rate of nitrification in the cathode chamber, so nitrification and denitrification can occur simultaneously (Feng et al., 2015; Park et al., 2009). Another advantage of this system is that it does not require additional supplementation with alkali reagents, due to the generation of hydroxyl ions during the oxygen reduction reaction and the potential of denitrification to compensate for the reduction of alkalinity in nitrification.

Taking advantage of the merits of the combination of MFC and SND as we demonstrated previously (Feng et al., 2015), we herein aim to demonstrate the utilization of a single MFC reactor to accomplish real coking wastewater treatment. Both batch-fed and continuous-flow experiments were performed to examine the feasibility of simultaneous carbon and nitrogen removal from coking wastewater with zero alkaline consumption. The degradation kinetics of COD, volatile phenols, thiocyanate and TN were assessed, and the concentrations of the main compounds available in the coking wastewater, such as phenol, methylphenol, dimethylphenol, naphthol, quinoline, isoquinoline, pyridine, and indole, were determined before and after the one-step one-reactor treatment. The structure and composition of the microbial community were analyzed to identify the functional bacteria possibly responsible for the biodegradation of various components. Furthermore, the shifts in the microbial communities in response to different experimental conditions were investigated. To go beyond this demonstration, we attempt to establish the relationships of the COD and TN degradation rates with the current density, a critical factor affecting the degradation performance of single MFC.

2. Materials and methods

2.1. Coking wastewater sample

The raw coking wastewater was collected from the wastewater treatment plant of Shaoguan Steel Company (Guangdong, China), which has an average treatment capacity of 2000 m^3 per day and includes physicochemical pretreatments, biological treatments, and advanced oxidation treatments. During the physicochemical pretreatments, $FeSO_4$ was mixed with the wastewater to coagulate and precipitate the suspended solids, including CN^- and S^{2-} (Yu et al., 2016). The wastewater fed to the biological system contained the following primary components: 2801.3 ± 45.2 mg L^{-1} COD, 701.9 ± 20.2 mg L^{-1} volatile phenols, 250.5 ± 5.5 mg L^{-1} NH_4^+–N, 358.8 ± 12.2 mg L^{-1} thiocyanate, and 350.4 ± 7.9 mg L^{-1} TN. This wastewater was used as the sample for the following experiments and analyses.

2.2. MFC setup, inoculation, and operation

The two-chamber MFC was constructed according to the procedures described previously (Xie et al., 2014; Feng et al., 2015; Yu et al., 2017). The cathode and anode chambers were separated by an anion-exchange membrane (Zhejiang Qianqiu Group Co., Ltd. China), and each had a volume of 250 mL. The electrodes in both chamber were made of graphite felt (7.0 × 4.0 × 0.5 cm), which was connected to the external circuit with a titanium wire (1.0 mm in diameter). A resistor box combined with a voltage collector was used to connect the two electrodes. For inoculation, cultures taken from the anode and cathode of a running MFC were added respectively to each chamber, resulting in the production of electricity and the degradation of phenol and ammonium (Feng et al., 2015). The anolyte (pH 7.0) consisted of 0.25 g L^{-1} NH_4Cl, 2.94 g L^{-1} NaCl, 0.10 g L^{-1} KCl, 1.00 g L^{-1} $NaHCO_3$, 0.015 g L^{-1} $CaCl_2$, 1.00 g L^{-1} $Na_2HPO_4 \cdot 12H_2O$, 2.72 g L^{-1} $CH_3COONa \cdot 3H_2O$, 10 mL L^{-1} of mineral solution (Xie et al., 2014) and 10 mL L^{-1} of vitamin solution (Xie et al., 2014). The cathode chamber was fed with the same medium with the exception of acetate. In addition, synthetic wastewater containing ammonium and phenol was amended to the cathode chamber. To adapt the bacteria to the high loadings, the concentration of NH_4^+–N and phenol was increased gradually from 50 to 250 mg L^{-1} and from 500 to 2500 mg L^{-1}, respectively. During inoculation, the MFC was operated at 30 °C under the batch-fed mode and a resistor of 500 Ω was used to connect the anode and the cathode. The catholyte was pumped with air, with an initial dissolved oxygen (DO) concentration of 8.3 ± 0.5 mg L^{-1} in the chamber. When the MFC was capable of generating electricity and completely degrading ammonium and phenol, three quarters of the old catholyte and anolyte were replaced with fresh catholyte and anolyte, and the remaining liquid (50 mL) was used to inoculate the MFC.

After inoculation, which lasted over two months, the synthetic wastewater fed to the cathode chamber was replaced with the real coking wastewater and no other chemicals were added to the cathode chamber. The anolyte fed to the anode chamber remained the same. The MFC was first operated at a resistance of 500 Ω in batch-fed