



Regular article

Integration of microbial fuel cell and catalytic oxidation reactor with iron phthalocyanine catalyst for Congo red degradation



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ABSTRACT

An integrated system of a microbial fuel cell (MFC) and a catalytic oxidation reactor (COR) was developed. In the MFC–COR system, H_2O_2 generated at the MFC cathode and residual dissolved oxygen could both be used as oxidants for Congo red degradation in the presence of iron phthalocyanine-based catalyst. Over 90% of Congo red was degraded within 72 h in neutral solution. The open-circuit potential and maximum power density of the MFC reactor were measured as 0.615 V and 808.3 mW/m³, respectively. The iron phthalocyanine-based catalyst was still active enough even after 33 cycles of operation. LC–MS analysis showed that Congo red was decomposed into less toxic and more biodegradable organics including malonic acid and maleic acid. This MFC–COR system can be a promising alternative for simultaneously biodegradable and refractory pollutant removal.

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1. Introduction

Microbial fuel cell (MFC) is an innovative energy-converting system that can produce electricity directly from organic or inorganic matter oxidation [1,2]. In such a device, electro-active microbes acting as biocatalyst extract electrons from substrate and transfer them onto the anode. These electrons then travel through an external circuit to the cathode, where they react with the final electron acceptors (such as oxygen or nitrate). A wide range of biodegradable substances, such as glucose, proteins and domestic wastewaters [3], can be used as fuels in this process, making it a promising technology for wastewater treatment.

Usually, oxygen is supplied to the cathode of MFC and mainly converted to water via a four-electron pathway to gain more electric power, but noble platinum is often essential [4]. Oxygen can also be reduced via a two-electron process on carbon-based electrodes, then producing a valuable chemical, hydrogen peroxide (H_2O_2) [5]. In the presence of ferrous ion under acidic condition, H_2O_2 is activated to generate hydroxyl radicals ($\cdot OH$), which is known as the Fenton reaction. This MFC–Fenton system can utilize the chemical energy of biodegradable compounds in the anode chamber to

degrade bio-refractory pollutants in the cathode chamber [6,7]. The homogeneous Fenton process, however, suffers from intensive iron sludge production, which requires extra treatment and management. Heterogeneous catalysts like γ -FeOOH [8] and limonite [9] have been used to minimize the consumption of catalyst. These iron-based catalysts can only utilize the generated H_2O_2 to oxidize organic pollutants, but excess dissolved oxygen was failed to be made full use of.

Iron phthalocyanine (FePc) resembles the active sites of enzymes responsible for catalytic aerobic oxidations and destruction of peroxides and it can activate both H_2O_2 and O_2 to degrade recalcitrant organic pollutants [10,11]. The catalytic process involves a nucleophilic iron(III) peroxy complex and following conversion into high-valent metal-oxo compounds as the active species, which is highly different from the Fenton system [12]. Water-soluble FePc compounds are effective in wastewater treatment, but they are difficult to be separated from the effluents for reuse, thereby causing a secondary contamination and catalyst loss. Recently, some solid materials, such as cellulosic fiber [13], chitosan [14] and carbon nanotubes [15], have been employed as supporter to avoid the defects of water-soluble FePc compounds for recalcitrant pollutant degradation. And recalcitrant pollutants, such as trichlorophenol [16], dyes [13] and chelating agents [17], can be degraded to organic acids with low molecule weight and even mineralized into carbon dioxide. However, these studies are mainly

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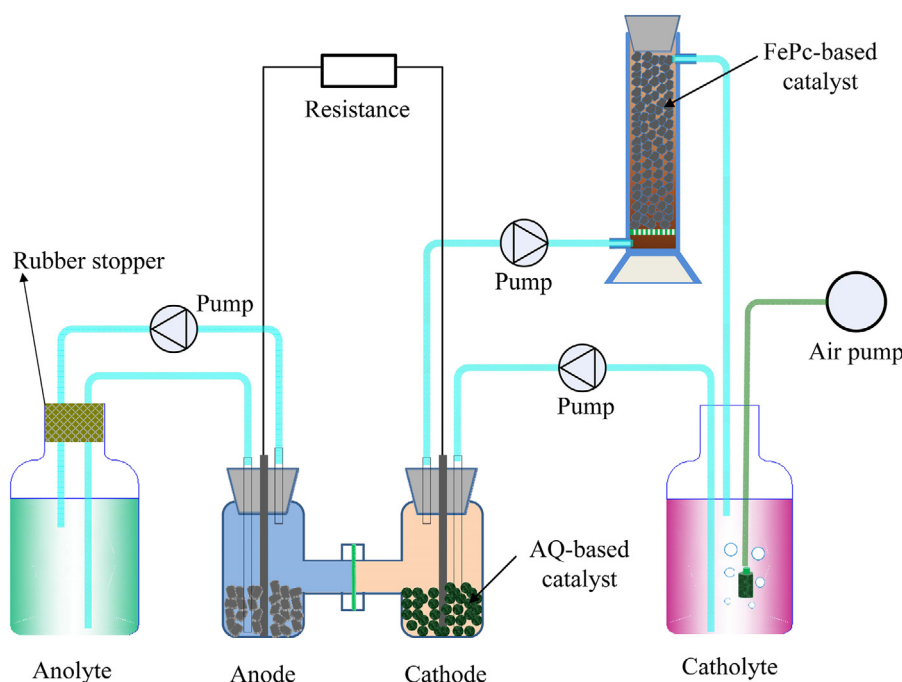


Fig. 1. Construction of the MFC-COR system.

focused on using H_2O_2 as the oxidant and little attention has been paid to FePc/ O_2 system for decontamination.

In our previous study, anthraquinone monosulfonate adsorbed on the basal plane of reduced graphene oxide was found showing a good electrocatalytic activity towards oxygen reduction reaction and the products was mainly H_2O_2 at low overpotential [18]. In another work of our group, leafy graphite nanosheet anchored FePc nanorods was synthesized and showed best performance in organic dye degradation with H_2O_2 in neutral solution [19]. In the present work, an integrated system of a MFC reactor and a catalytic oxidation reactor (COR) filled with granular catalyst (MFC-COR system) was developed to degrade a typical azo dye, Congo red. In this system, glucose was used as substrate for electroactive microbes in the anode chamber and O_2 was reduced via a two-electron process in the cathode chamber to generate H_2O_2 . These H_2O_2 molecules accompanying with residual dissolved oxygen were delivered into the COR reactor to react with catalyst for decolorization. To reduce the usage of FePc, it was firstly supported on exfoliated graphite (EG) and then bonded with polytetrafluoroethylene (PTFE). This composite material was used as cheap catalyst in COR. The performances in decolorization and electricity production of the integrated system were explored, and the stability of the catalysts was evaluated after 33 cycles of operation. The degradation products of Congo red were analyzed by liquid chromatography-mass spectrometry (LC-MS) technology, and finally electron transfer mechanisms of the MFC-COR system were proposed.

2. Experimental

2.1. Construction of the MFC-COR system

The MFC-COR system was mainly composed of an H-type MFC, a COR reactor and some auxiliary facilities, such as aerating apparatus, tanks for electrolytes, peristaltic pumps and some rubber hose, as seen in Fig. 1.

A two-chamber bottle-type MFC made of glass was used in this study, and each chamber has an internal diameter of 4.0 cm and a

height of 8.0 cm. The effective volume of each chamber was 100 mL and the two compartments were separated by a proton exchange membrane (Nafion 117, Dupont) with a cross-section diameter of 1.2 cm. Granular graphite (diameter 6–8 mm) and a graphite rod (8 mm in diameter) were used as the anode, which accounted for approximate 30% of the liquid volume in the anode chamber. Some anthraquinone (AQ)-based spherical catalyst (diameter 3–5 mm) and a graphite rod (8 mm in diameter) were used as the cathode, which also occupied approximate 30% of the liquid volume in the cathode chamber. The AQ-based catalyst was mainly composed of graphite particles, reduced graphene oxide (RGO), anthraquinone-2-sulfonic acid and PTFE, and its preparation method was presented in Supplementary Section S1. The anode and cathode chambers were all sealed with rubber stoppers.

A polymethyl methacrylate cylinder with a diameter of 2.9 cm and a height of 14 cm was used as the COR reactor. The FePc-based catalyst (Prepared as Supplementary Section S2) occupying 42 mL of the liquid volume was filled into the reactor and then sealed with a rubber stopper. The cylindrical reactor was covered with aluminum foil to exclude the effect of visible light radiation.

2.2. Operation of the MFC-COR system

The MFC anode was inoculated with pre-acclimated microbial community from another MFC with glucose used as substrate that had been operating for more than two years, and the culture medium contained glucose (1.5 g/L), NaHCO_3 (0.25 g/L), NH_4Cl (0.3 g/L), KCl (0.13 g/L), phosphate buffer solution (PBS, 0.02 M, pH = 7.0) and some trace elements. Dissolved oxygen was removed from the medium by sparging the solution with nitrogen gas for at least 20 min before use. The anolyte of 1.0 L was recirculated by a peristaltic pump at a flow rate of 0.12 L/h to maintain constant reducing power. The catholyte of 0.5 L composed of 0.1 M PBS (pH = 7.0) and 100 μM Congo red was pumped into the cathode chamber at a flow rate of 2.24 L/h, passed through the COR reactor and then returned to the tank. Throughout all experiments, the anolyte was sealed with a rubber stopper to prevent the air into the solution, while the catholyte was bubbled with an air

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