



Improvement of anaerobic biological treatment effect by catalytic micro-electrolysis for monensin production wastewater

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HIGHLIGHTS

- Novel catalytic-ceramic-filler (CCF) prepared from solid waste (scrap iron) and clay.
- Catalytic micro-electrolysis (CME) with CCF used as monensin wastewater pretreatment.
- About 98.44% of monensin residue and 37.07% of COD were removed by the CME reactor.
- The UASB efficiency (methane yield and COD removal) improved by the CME pretreatment.
- The coupled CME-UASB-AS system removed about 98% of COD and 95% of chroma.

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ABSTRACT

Monensin discharged with the animal wastes and wastewater can cause harmful effect to the environment and human health. In this study, catalytic micro-electrolysis (CME) reactor filled with novel catalytic-ceramic-filler was utilized as pretreatment to improve the anaerobic biological treatment effect for the real monensin wastewater. The CME reactor as a possible pretreatment process had satisfactory effect, with 98.44% of monensin residue and 37.07% of chemical oxygen demand (COD) removals at the optimum hydraulic retention time (HRT) of 3.0 h and dissolved oxygen (DO) of about 1.5 mg L⁻¹. Subsequently, as the secondary biological treatment, the Up-FLOW Anaerobic Sludge Blanket (UASB) reactor treatment effect was greatly improved by the CME pretreatment, with approximately 80% of COD removal at the optimum organic loading rate (OLR) of 3.5 kg m⁻³ d⁻¹, which had higher methane yield (about 0.33 m³ kg⁻¹ COD⁻¹) and lower volatile fatty acids (VFA) concentration (about 300 mg L⁻¹) than that of the UASB reactor without pretreatment. Finally, an activated sludge (AS) reactor was utilized as the last biological treatment and the coupled CME-UASB-AS system had high COD and chroma removal (about 98% and 95%, respectively), the final effluent (COD and chroma of about 200 mg L⁻¹ and 40, respectively) with no residual monensin met the national discharged standard, which provided a reliable system for the practical monensin production wastewater treatment.

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1. Introduction

Monensin, as a kind of polyether ionophoric antibiotics, has been widely used in animal feeding operations [1]. It is usually added to ruminant diets to improve the efficiency of feed utilization, and may be subsequently released to the environment with animal wastes through overflow or leakage from storage structures or land application [2,3]. Moreover, monensin productive process usually generates large amount of wastewater, which contains

high concentration of monensin residue. Monensin derived from streptomyces species has been considered as high risk compound, which comprises of complex molecules with high molecular weight [4]. Whether animal wastes or the wastewater is discharged, the contained monensin will cause harmful effect to the environment and human health. Therefore, it is significant to dispose of the monensin production wastewater to prevent environmental pollution from the discharged monensin.

Catalytic micro-electrolysis (CME), based on traditional micro-electrolysis, has been attracted much attention from researchers in the past several years, probably due to its advantage in refractory wastewater treatment. It can improve traditional micro-

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Nomenclature

ME	micro-electrolysis	LC ₅₀	lethal concentration of 50%
CF	ceramic filler	CME	catalytic micro-electrolysis
HRT	hydraulic retention time (h)	CCF	catalytic-ceramic-filler
DO	dissolved oxygen (mg L ⁻¹)	UASB	Up-flow Anaerobic Sludge Blanket
AS	activated sludge	COD	chemical oxygen demand (mg L ⁻¹)
BOD	biochemical oxygen demand (mg L ⁻¹)	VFA	volatile fatty acids (mg L ⁻¹)
EC ₅₀	concentration for 50% of maximal effect		

electrolysis, lower concentrations of toxic components in aqueous solution and enhance the biodegradability of wastewater. Fe/Cu bimetal system, as a kind of CME, can accelerate corrosion of Fe⁰ and hence improve the micro-electrolysis ability. Therefore, Fe/Cu catalytic micro-electrolysis has been utilized to remove many pollutants, such as hexavalent chromium [5], nitrate [6], indigo blue [7], and trichloroethene [8], etc. It is well known that filler plays a key role in a CME system, which determines the treatment efficiency and practical application of CME. Therefore, a new type of cost-effective and easily applicable filler is urgently needed to overcome disadvantages of traditional fillers (short-circuiting and clogging during operation) and improve the Fe/Cu CME technology.

As a kind of anaerobic biological treatment technology, Up-flow Anaerobic Sludge Blanket (UASB) was firstly invented by Professor G. Lettinga in 1977 [9], and characterized by high organic and hydraulic loading, short hydraulic retention time, high active biomass concentrations, easily controlling and operating, energy- and cost-efficient, etc [10]. When applied in the wastewater treatment, a UASB system usually degrades the organic pollutants through four stages in turn, including hydrolysis, acidogenesis, acetogenesis, and methanogenesis, and the organic pollutants are finally transformed into methane and carbon dioxide [11,12]. At present, UASB has been widely applied in high concentrated organic wastewater treatment, including textile wastewater [13], 3,4,5-trimethoxybenzaldehyde and Di-bromo-aldehyde manufacturing wastewater [14], cheese whey wastewater [15], chloronitrobenzenes wastewater [16], potato-juice wastewater [17], palm oil mill effluent [18], etc. Thus, UASB is a cost-effective and high-efficiency technology for treating the industrial wastewater with high concentration.

In this study, a coupled CME-UASB-AS system was applied in the real monensin production wastewater treatment. Firstly, the prepositive CME system was utilized as the pretreatment to remove the monensin residue in the wastewater, which could reduce the negative effect of monensin on the anaerobic system (UASB) and enhance the treatment effect of the UASB system. Subsequently, the later biological treatments (UASB + AS) were applied to degrade the organics after the CME pretreatment, which could completely and effectively dispose of the real monensin production wastewater. Therefore, the objectives of this study were to: (1) fill novel catalytic-ceramic-filler (CCF) in a catalytic micro-electrolysis (CME) system for monensin production wastewater pretreatment and determine the optimum operating conditions for the CME system; (2) investigate the influence of CME pretreatment on methane yield and chemical oxygen demand (COD) removal by a Up-flow Anaerobic Sludge Blanket (UASB) system for the secondary wastewater treatment and determine the optimum operating conditions for the UASB system with the CME pretreatment; (3) couple the CME, UASB and aerobic activated sludge (AS) system for the monensin production wastewater systematic treatment and make the final effluent meet the requirement of the national discharged standard (COD ≤ 300 mg L⁻¹ and chroma ≤ 60, C standard of CJ 343-2010, China).

2. Materials and methods

2.1. Preparations of fillers

Ceramic filler (CF) and catalytic-ceramic-filler (CCF) were prepared for micro-electrolysis (ME) and catalytic micro-electrolysis (CME) respectively according to our previous study [19]. Clay (obtained from the mountainous area in Zibo city of Shandong Province, China) and scrap iron (obtained from a machinery plant in Jinan city of Shandong Province, China) were utilized to prepare ceramic filler (CF) for the ME reactor, while copper sulfate (CuSO₄·5H₂O, purchased from Sigma-Aldrich, St. Louis, MO) solution (Cu²⁺ of 5.0 g L⁻¹) was added to sinter the catalytic-ceramic-filler (CCF) for the CME reactor. The chemical components of the clay are shown in Table S1, and the curved scrap iron (4.0–5.0 mm wide; 1.0 mm thick) had an average specific surface area of 0.13 m² g⁻¹.

Firstly, the clay and scrap iron were oven dried at 105 °C for 4.0 h, crushed in a ball mill and sieved (the diameter of the sieve mesh was 0.154 mm). Subsequently, clay and scrap iron (3:2, w/w) were stirred in a dry powder stirrer for 10 min, followed by pouring into a pelletizer (DZ-20) to produce pellets (10.0% water added for CF; 10.0% copper sulfate solution added for CCF). Then, the raw pellets were sieved (the diameters were 5.0–6.0 mm) and stored in draught cupboard at room temperature (22 °C) for 24 h. Secondly, the dried pellets were rapidly transferred into an electric tube rotary furnace (KSY-4D-16) and sintered at 850 °C for 30 min in nitrogen atmosphere. Finally, the sintered pellets were kept in a vacuum drying oven to cool down to room temperature (22 °C).

The appearance and microstructure of CF and CCF (Fig. S1) showed that CF and CCF had rough surfaces and porous frameworks, which might increase their specific surface area, improve water flow and enhance water mass transfer, resulting in the improvement of pollutants removal efficiency by the reactors packed with CF and CCF. Additionally, the results of the leaching tests of CF and CCF (Table S2) showed that the concentrations of all the detected metals (Cu, Zn, Pb, Cr, Cd, Hg, Ba, Ni, and As) in the lixivium were much lower than the limits of the national standards (GB 5085.3-2007, China, Hazardous Wastes Distinction Standard-Leaching Toxicity Distinction), revealing that utilization of CF and CCF would not cause harm to the water environment.

2.2. Reactors for the treatment system

A pilot-scale system as shown in Fig. 1, consisting of five identical cylindrical columns made of polypropylene, was set up for the monensin production wastewater treatment. The first two reactors, with the diameter of 60 cm, height of 1.8 m and effective volume of 297 L, were placed in parallel to each other and used as the ME (packed with CF) and CME reactor (packed with CCF), respectively. From bottom to top, both ME and CME reactor were filled

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