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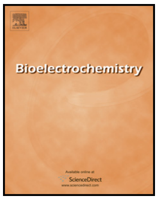

Nitrate removal from pharmaceutical wastewater using microbial electrochemical system supplied through low frequency-low voltage alternating electric current

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ABSTRACT

In this study, a microbial electrochemical system (MES) was designed to evaluate the effects of a low frequency-low voltage alternating electrical current on denitrification efficacy in the presence of ibuprofen as a low biodegradable organic carbon source. Cylindrical carbon cloth and stainless steel mesh electrodes containing a consortium of heterotrophic and autotrophic bacteria were mounted in the wall of the designed laboratory-scale bioreactor. The effects of inlet nitrate concentration (50–800 mg L^{-1}), retention time (2.5–24 h), waveform magnitude (0.1–9.6 V_{p-p}), adjustable direct current voltage added to offset voltage (0.1–4.9 V), alternating current frequency (10–60 Hz), and waveforms (sinusoidal, square, and ramp) were studied in this work. The results showed that the proposed system removes 800 mg L^{-1} nitrate up to 95% during 6.5 h. Optimum conditions were obtained in the 8 V_{p-p} using a frequency of 10 Hz of a sinusoidal waveform. The morphology studies confirmed bacterial morphology change when applying the alternating current. Dehydrogenase activity of biofilms formed on surface of stainless steel electrodes increased to 15.24 μgTF $mg_{biomass}$ cm^{-2} d. The maximum bacterial activity was obtained at a voltage of 8 V_{p-p}. The experimental results revealed that the MES using a low frequency-low voltage alternating electrical current is a promising technique for nitrate removal from pharmaceutical wastewaters in the presence of low biodegradability of carbon sources such as ibuprofen.

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1. Introduction

Due to anthropogenic activities, some industrial and agricultural sources discharge their wastewater containing high nitrate and low organic biodegradable into the environment [1]. Although various techniques have been proposed for the simultaneous removal of nitrate and organic compounds, they are not free from drawbacks [2]. Some industrial processes generate wastewater with high levels of nitrate and organic compounds that must be removed simultaneously [3]. For example, wastewater of pharmaceutical factories has high levels of toxic, complex, and poorly biodegradable compounds accompanied by nitrogenous compounds [4]. The toxicity of the organic compounds on microorganisms is a major problem during biological treatment [5]. The type of organic material used as a carbon source for heterotrophic denitrification is the main parameter affecting nitrate removal [6]. To increase denitrification efficiency, various methods have been proposed; e.g., addition and tuning of nutrients and electron donor/acceptor ratio, electrochemical hydrogen production for autotrophic denitrifying bacteria, and enhancement of bacteria activity by electrostimulation [1,7,8]. Available data show that the induced electrical current can stimulate bacteria metabolism and enhance biochemical performance [9–11]. For example, Hao et al. reported a single chamber microbial fuel cell and a bioelectrical reactor for vanadium removal. The results showed that the highest removal efficiency of vanadium was 93.6% at 12 h operation. Recently, microbial electrochemical systems (MESs) have been proposed for bioremediation of various environmental pollutants. MESs refer to a set of biological systems in which bacteria catalyze the oxidation or reduction reactions [12]. Electric currents can be divided into two branches: alternating current and direct current. Applying direct current to electrochemical systems can produce highly active chemical byproducts such as •OH, O_3, H_2O_2, and Cl_2 [13]. So, the current in bioelectrochemical systems can lead to toxic byproducts for microorganisms [14]. In the alternating current, on the other hand, the magnitude of frequency changes periodically while net charge is zero. Utilizing a low voltage-low-frequency with an alternating current may solve some problems such as anode corrosion and cathode inactivation [15]. Previous studies have shown that applying a low voltage-low frequency alternating electric current in a bioreactor can create several changes including dense and fast-settling sludge granules, increase in enzyme activity, changes in morphological and biochemistry

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characteristics [16]. It has been reported that biomass activity could be enhanced as a result of alternating current utilization. In the present study, a low voltage very low frequency alternating current at different waveforms was applied on bacteria community as electrostimulation. The effect of inlet nitrate concentration, retention time (h), the magnitude of a waveform [AMPL / V_{p-p}], an adjustable DC voltage added to the signal output [OFST/V], alternating current frequency, and waveform was studied on the biological denitrification process. Several studies have been conducted on electrostimulation of bacteria using direct current [13]. However, to the best of our knowledge, rare studies have rarely reported on low voltage very low-frequency alternating currents to enhance biological activities. It is expected that experimental results could be used as a reference for applying MES through a low frequency-low voltage alternating electrical current for biological denitrification in industrial wastewater treatment.

2. Materials and methods

2.1. Materials and bioreactor configuration

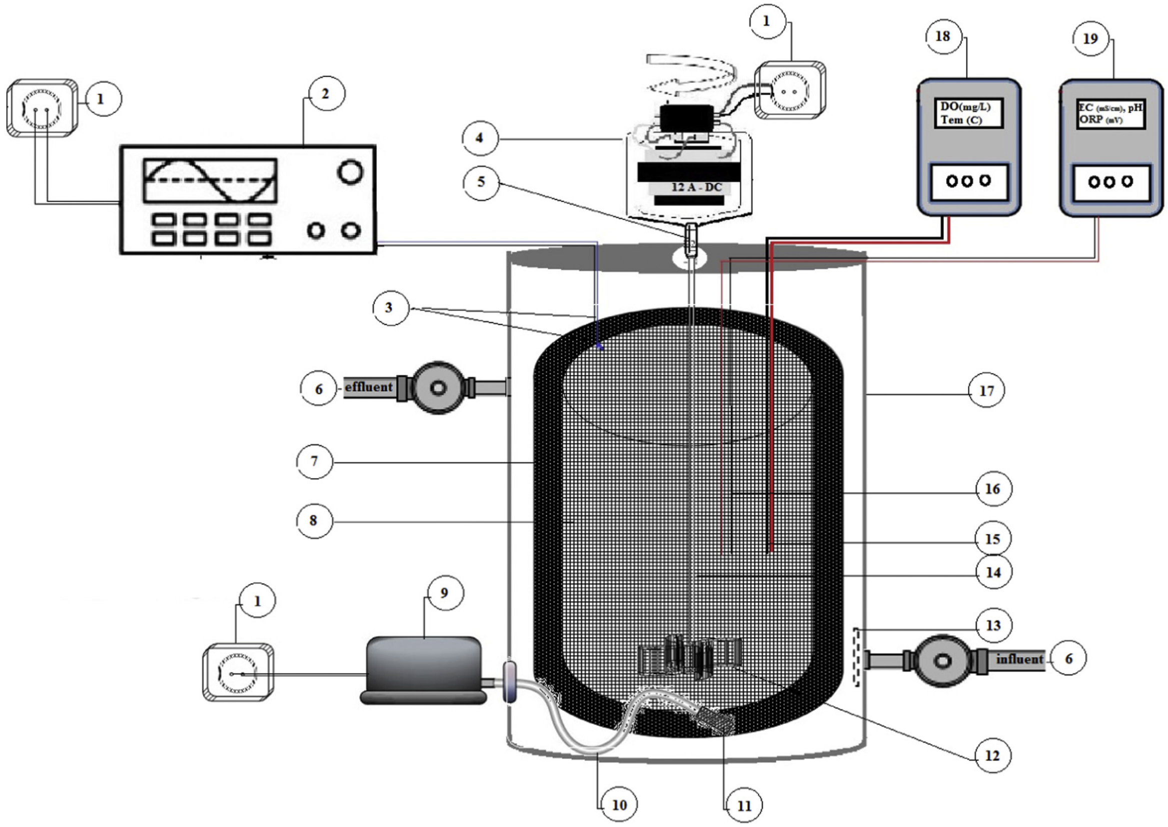
The experimental bioreactor consisted of a glass vessel with 5 L effective capacity. Cylindrical carbon cloth and stainless steel mesh with 2 cm inter-electrode distance were mounted in the wall of the bioreactor. The bioreactor was stirred manually to ensure complete mixing. The MES configuration is shown in Schematic 1. Nutrients including potassium nitrate, ibuprofen, KH_2PO_4, and K_2HPO_4 were inoculated in the reactor at a C:N:P ratio of 3:1:0.2 for microbial growth [17,18]. The chemical structure of ibuprofen is shown in Fig. 1. The pH of the influent synthetic wastewater was adjusted to 7.3 ± 0.3 before inoculating the bioreactor. All chemicals used in this study were of analytical reagent grade and used without further purification. Potassium nitrate and ibuprofen were used as nitrate and carbon sources, respectively.

2.2. Experimental procedure

The inoculum for growing the denitrifying microorganisms was obtained from a wastewater treatment plant, Tehran, Iran. The concentration of the suspended biomass added to the bioreactor was 4000 mg L^{-1} as a mixed liquor suspended solid (MLSS). The ratio of Volatile Suspended Solids (VSS) to Total Suspended Solids (TSS) was 82%. Initial electrical inputs were AMPL = 8 V_{p-p}, OFST = 0.2 V, waveform = sinusoidal, frequency = 10 Hz, and duty cycle = 75%. The term peak-to-peak voltage (V_{p-p}) refers to a maximum alternating current for the function generator. After formation of a biofilm, the reactor contents became gray in color, and effluent turbidity dropped after 14 days. Subsequently, nitrate was added to the bioreactor at various concentrations (50–600 mg L^{-1}) with a theoretical C/N ratio of 4.2 using ibuprofen as a carbon source. The start-up was completed after nitrate reduction efficiency was approximately 90% in five-time intervals. The bioreactor was operated for 45 days. Dissolved oxygen was maintained at <0.5 mg L^{-1} and all experiments were operated at 25 ± 2 °C. Operational conditions (retention time, C/N, AMPL, OFST, and frequency) were controlled in constant mode to attain steady-state denitrification.

2.3. Analytical methods

All effluent samples were filtered with a 0.45 μm pore size Whatman cellulosic paper filter before analysis. Ammonia and nitrite were analyzed according to the standard methods [19]. Nitrate was determined using a spectrophotometer at λ_{max} 220 nm and 275 nm, according to a standard APHA method. Chemical Oxygen Demand (COD) was also



Schematic 1. The configuration of the MES; 1) timer; 2) function generator; 3) function generator to electrode connector; 4) electromotor; 5) flexible shaft coupling; 6) peristaltic pump; 7) carbon cloth electrode with mesh plastic support; 8) stainless steel electrode; 9) air pump; 10) air transfer tube; 11) porous rock; 12) mixing blade; 13) perforated baffle; 14) shaft; 15) dissolved oxygen meter probe; 16) electrical conductivity, oxidation-reduction potential, and pH meter probe;17) glass vessel of the bioreactor; 18) DO meter; and 19) EC, ORP, and pH meter.

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