



Coupling of aerobic/anoxic and bioelectrogenic processes for treatment of pharmaceutical wastewater associated with bioelectricity generation



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ABSTRACT

A sequential treatment strategy designed by integrating sequencing batch (anoxic/aerobic operation) reactor (SBR) with bio-electrochemical treatment (BET) was studied to enhance the remediation of real-field pharmaceutical wastewater (PW). Study was carried out by feeding PW to two SBR systems operated under aerobic (SBR_{Ae}) and anoxic (SBR_{Ax}) microenvironments. Comparatively higher substrate degradation (SD) and multi-pollutant removal was observed with SBR_{Ax} (68.69%) in comparison to SBR_{Ae} (60.27%), due to the switching of bacterial metabolism that facilitates redox reactions. In order to further enhance the treatment efficiency, the effluents resulting from SBR_{Ax} were fed to BET₁ and SBR_{Ae} to BET₂. Relatively higher bioelectrogenic activity and SD were exhibited by BET₁ (Voltage: 536 mV; current: 1.21 mA; SD: 75%) than BET₂ (Voltage: 323 mV; current: 2.67 mA; SD: 73%). Self-induced bio-potential developed in BET system due to electrode assembly enabled higher organic and inorganic compounds removal than SBR. Study illustrated the advantage of integration strategy in enhancing the treatment of PW with simultaneous bioelectricity generation.

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1. Introduction

Pharmaceutical industries use an array of organic, inorganic and biological reactions for the synthesis and formulation of drugs for which copious quantities of water is being utilized [1–3]. Manufacturing process from these industries discharge wastewater, which are recalcitrant in nature and contains inorganic salts. Various methods such as membrane distillation, fenton oxidation, photocatalysis, ultrasound radiation, etc. are being used for the treatment of pharmaceutical wastewater (PW). However, these methods mandate additional cost and energy investments. Alternatively, biological methods are less energy intensive and are being widely used for wastewater treatment due to their sustainable nature [4–7]. Advanced biological wastewater treatment process such as sequencing batch reactor (SBR) is being employed to treat high strength wastewaters, as it facilitates multiple operations in a single system which eventually lead to improved treatment efficiency [8–10]. Operation under diverse microenvironments will increase the capabilities of biocatalyst to function effectively. SBR

process is considered to be advantageous due to the operational flexibility which easily allows varying the feeding regime in a single compact reactor [6,11–14].

On the other hand, microbial fuel cell (MFC) otherwise called as bioelectrochemical treatment system (BET) are emerging as potential process for multi-pollutants removal [15–21]. BET works on the basis of MFC principle which can facilitate the direct conversion of organic substrate present in wastewater to electricity through a cascade of redox reactions. Presence of electrode assembly in BET aids in the development of biopotential necessary for the breakdown of complex organic compounds and inorganic salts [22–28]. Besides, electrical energy can be harvested through anode and cathode chambers by linking microbial metabolism through electron donation and reduction.

Since SBR process can treat the complex wastewaters only upto certain extent, there is a need for further remediation to decrease the complexity of wastewater. In order to achieve this, SBR process can be coupled with BET system that enables the breakdown of inorganic salts and enhance the treatment of recalcitrant PW. Similar studies have been reported for enhanced treatment of petrochemical wastewaters and textile based wastewater [29,30]. Considering the advantage of both the processes, present study is

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designed to integrate SBR as a pre-treatment step to BET process to achieve maximum treatment of PW. The study was executed in two steps. In the first step, two SBR reactors (SBR_{Ax} and SBR_{Ae}) were operated with PW under anoxic and aerobic microenvironments respectively. The effluents resulting from SBR_{Ax} were fed into BET_1 and SBR_{Ae} to BET_2 to enhance the treatment efficiency. The performance of SBR and BET was comparatively assessed in terms of treatment and bio-electrochemical parameters.

2. Materials and methods

2.1. Bioreactors

SBR systems were operated under aerobic (SBR_{Ae}) and anoxic (SBR_{Ax}) microenvironments. Reactors were fabricated using acrylic material with a capacity of 1/0.99 L total/working volume (diameter 7.5 cm and length 22.5 cm). In SBR_{Ae} , oxygen was sparged continuously using an aerator, and in the anoxic reactor, microaerophilic condition was maintained by sparging air for 5 min for every 4 h interval. Single chamber bioelectrochemical system (BET) was designed and fabricated with a total/working volume of 1.1/1.0 L (dimensions 14 × 11 × 4 cm). In BET systems, stainless steel mesh (grade: 316L) was used as an anode with a projected surface area of 0.28 m² and non-catalysed graphite as cathode with a surface area of 0.24 m² and a distance of 3 cm between anode and cathode. Anode was completely submerged in the anolyte, whereas cathode was partially submerged (bottom portion) and top portion was exposed to atmospheric air. Proper provisions were made in all biosystems for feeding, decanting, recirculation and air supply operations. Real field pharmaceutical industry wastewater (PW) with COD of 29000 mg/l, TDS of 5442 mg/l, pH: 9.04; nitrates: 974 mg/l; phosphates: 138 mg/l; sulphates: 29.5 mg/l was used in the study. Prior to use, effluent was stored at 4 °C. Before feeding, pH of PW was adjusted to 7.0 ± 0.2 using 0.1 N HCl.

2.2. Biocatalyst

Aerobic consortia procured from a full-scale effluent treatment plant (ETP) was used as parent culture for both SBR_{Ae} and SBR_{Ax} . The consortia were enriched by sparging air for 5 min with an interval of every 4 h. In the case of BET, anaerobic consortium from a full-scale anaerobic effluent treatment plant was used as parent inoculum. Parent cultures were washed twice with saline buffer (5000 rpm, 20 °C) and enriched in designed synthetic wastewater [DSW; glucose – 3 g/l; NH_4Cl – 0.50 g/l, KH_2PO_4 – 0.25 g/l, K_2HPO_4 – 0.25 g/l, $MgCl_2$ – 0.30 g/l, $CoCl_2$ – 25 mg/l, $ZnCl_2$ – 11.50 mg/l, $CuCl_2$ – 10.50 mg/l, $CaCl_2$ – 5 mg/l, $MnCl_2$ – 15 mg/l, $NiSO_4$ – 16 mg/l, $FeCl_3$ – 25 mg/l] at required microenvironments.

2.3. Experimental design and operations

Raw effluent of PW was fed to SBR and the resulting effluent was fed to BET to treat the residual waste. SBR operation was carried out in two different microenvironments, viz., aerobic (SBR_{Ae}) and anoxic (SBR_{Ax}) with an organic loading rate of 12.08 kg COD/m³day with sequential operation comprising of 15 min of filling phase (FILL), 2820 min of recirculation (REACT) phase, 30 min of settling phase (SETTLE) and 15 min of decant phase (DECANT). The outlet of SBR reactors was subsequently fed into BET systems viz., BET_1 (SBR_{Ax} effluent) and BET_2 (SBR_{Ae} effluent). Bioreactors were operated in suspended growth configuration with a cycle period (retention time) of 48 h at ambient room temperature.

2.4. Analytical procedures

The performance of SBR and BET systems with respect to treatment efficiency was evaluated by monitoring chemical oxygen demand (COD; 5220-C) (closed-reflux (titrimetric) method), sulphates (4500-E), nitrates (4500-B), phosphates (4500-D), pH (4500-H + B), and total Dissolved Salts (TDS) (2540 C) according to standard methods [31]. Voltage and current were measured using a digital multi-meter at 100 Ω resistance. Anode potential was measured with reference to Ag/AgCl(s) using anode as working electrode at varying resistances (30–0.05 kΩ). Polarization was performed across 30 to 0.05 kΩ varying resistances, during the stabilized phase of operation. Cyclic voltammetry (CV), linear sweep voltammetry (LSV) and chronoamperometry (CA) were used to study the bioelectrochemical behavior of biocatalyst using a potentiostat-galvanostat system by applying a potential ramp (+0.5 to –0.5 V) (Bio-logic-VSP). The assays in the BET system were performed considering anode (stainless steel mesh) as working electrode (WE) and cathode (graphite) as counter electrode (CE) against Ag/AgCl(s) reference electrode (RE). Voltammetry was performed for SBR considering anode (glassy carbon rod) as working electrode (WE) and cathode (platinum wire) as counter electrode (CE) against Ag/AgCl(s) reference electrode (RE).

3. Results and discussions

3.1. Substrate degradation

Initially, SBR_{Ae} and SBR_{Ax} were operated with DSW to facilitate the growth of biomass and subsequently, reactors were fed with real field PW. During initial cycles of operation, SBR_{Ae} (43.9%) and SBR_{Ax} (45.35%) reactors depicted marginal variation with respect to substrate removal efficiency, since both the reactors were inoculated with aerobic consortia. With the course of operation, anoxic reactor (68.96%) depicted higher substrate degradation compared to aerobic operation (60.27%) (Fig. 1a). In aerobic process, most of the chemical compounds are converted to their respective intermediate compounds via aerobic metabolism (oxidation reactions) and the electrons generated are accepted by oxygen as a terminal electron acceptor (TEA) thus limiting reduction reactions [32]. While in the case of anoxic process, since oxidation and reduction reactions occur alternatively, the chemical compounds are converted to their intermediates (oxidation), which are further simplified during the reduction reactions, thus enhancing treatment efficiency. Biocatalyst in the anoxic microenvironment might also depict a shift in metabolic function towards facultative in nature, due to the switching between alternate aerobic and anaerobic conditions, facilitate enhanced degradation of PW in SBR_{Ax} in comparison to SBR_{Ae} [32].

Despite the good COD removal efficiency obtained in the SBR process, complex intermediates and certain untreated compounds still persist in the effluents generated from SBR. Conversion of these compounds to simpler forms can be accomplished in BET, making SBR a pre-treatment process for pharmaceutical wastewater treatment. Therefore, in the present study, the effluents of SBR reactors with residual COD of 3.75 kg COD/m³day (SBR_{Ax}) and 4.8 kg COD/m³day (SBR_{Ae}) were subsequently fed to BET systems to further enhance substrate degradation. BET reactors also were initially operated with DSW for effective biomass growth and acclimatization of the biocatalyst to simple substrate. In comparison to both BET reactors, BET_1 showed treatment efficiency (75%) than BET_2 (73%). In comparison to SBR, BET system showed higher performance due to the presence of solid electrodes, which acts as an electron acceptor and develop a potential gradient aiding the cleavage of pharmaceutical residual organic compounds [21,23,29].

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