

Sequential nitrification and denitrification in a novel palm shell granular activated carbon twin-chamber upflow bio-electrochemical reactor for treating ammonium-rich wastewater

Introduction

The uncontrolled discharge of nitrogen components through nitrogen-rich wastewaters has been considered as a worldwide human health threat and environment challenge (Chen et al., 2006). In wastewater treatment plants with an anaerobic digester, reject water from dewatering digested sludge contains a significant amount of nitrogen (15–25% of the total nitrogen load in influent), which the effluent recycles to the head of the sewage treatment works (Mata-Alvarez, 2002; Dosta et al., 2007). The specific characteristics of the reject water, such as high concentration of ammonium and low C/N ratio, allow the application of special treatment methods. A separate treatment of reject water would significantly increase the elimination of nitrogen by decreasing the nitrogen load in the main stream (Fux et al., 2002).

Biological methods are more cost effective compared with physical–chemical techniques. In a conventional approach, nitrogen elimination is done through sequential nitrification and heterotrophic denitrification. The low ratio of C/N in reject water and high operational costs, attributed to O₂ provision and organic carbon necessities, have impeded broad implementation of conventional nitrification and denitrification. Recently, new methods, such as biological nitrogen removal via nitrite, have been applied as effective and economical approaches in treating reject water (Jenicek et al., 2007). Biological nitrogen removal is performed via nitrite due to the lower oxygen requirement (25%) and chemical oxygen demand (COD) 40% for denitrification, as well as faster kinetic of the process, in contrast with removal via nitrate. In addition, heterotrophic denitrification is deemed unfavorable (Zhao et al., 2011) due to limitations such as residual carbon sources, by-products that incur further post-treatment costs, clogging in reactors, and increases the effluent turbidity because of high growth rate. Therefore, researchers have used autotrophic bacteria; using inorganic compounds as carbon sources (CO₂ and NaHCO₃), is less expensive compared with using organic sources and has harmless residuals. On the other hand, autotrophic bacteria use inorganic compounds, such as manganese, hydrogen sulfide, ferrous iron, and molecular hydrogen as energy sources

(Ghafari et al., 2008; Mousavi et al., 2012). Among these compounds, H₂ is a perfect alternative to alleviate the previously mentioned problems due to its cleanliness. In addition, H₂ is the best electron donor among other elements (Mousavi et al., 2012). Consequently, bacteria that use hydrogen as energy source, namely, "autohydrogenotrophic bacteria," have been considered for the denitrification process. The application of autohydrogenotrophs in reactors requires proper methods of supplying hydrogen to the microorganisms. The initial injection of hydrogen gas into the reactor is a method with certain limits (Lee and Rittmann, 2002), such as low solubility of H₂ in water (1.6 mg L⁻¹ at 20 °C). This limitation declines the accessibility and raises the contact time for the biological process. Consequently, this also causes a decrease in the denitrification rate compared with heterotrophic denitrification (Prosnansky et al., 2002; Vasiliadou et al., 2006). Some marginal concerns are also reported in the application of hydrogen, such as the costly supply of H₂, flammability, as well as transport and storage problems (Ghafari et al., 2008). Thus, in situ hydrogen generation (water electrolysis) is recommended as an efficient technique to overcome these drawbacks. Therefore, researchers have considered a new integrated system namely bio-electrochemical reactor (BER) to treat water and wastewater.

A BER is defined as a system that consists of integrated processes (biological and chemical processes), which by electrolysis of aqueous solution prepare desirable oxic and anoxic conditions for microorganisms by passes the electric current through electrodes due to the production of suitable electron donors (H₂) and electron acceptors (O₂) as expressed in Eqs. (1) and (2) (Ramesh and Goel, 2005).

Please refer to the full text

A comprehensive review, recently by authors showed mainly previous studies have focused on eliminating nitrate via BERs (Mousavi et al., 2012) whereas, some researchers have applied this system for simultaneous or sequential nitrification and denitrification (SND) in different configurations. Watanabe et al. (2002) studied the possibility of simultaneous nitrification and denitrification in a single bio-electrochemical reactor. The single reactor enhances aerobic degradation by the production of oxygen in the anode zone, but adversely influences the anaerobic microorganism (Watanabe et al., 2002). Therefore, researchers have tried to establish a new configuration of BER as divided cell, which prepare the overall capacity of a BER without harmful effect of generated O₂ in anode on anoxic condition in cathode zone (Szekeress et al., 2001; Ramesh and Goel, 2005). In addition, researchers have attempted to speed up the biological treatment by finding ways to provide

better contact between the substrate and microorganisms via the immobilization of the biomass at the surface of the electrodes. The granular activated carbon (GAC), due to many advantages such as low cost, durable packing material, enhancement of the adhesion of bacteria and large macropore volume (Moreno-Castilla et al., 2003; Jiang and Li, 2009), has been applied in different studies as biocarrier and electrode to provide the surface for the creation of biofilm and increase the oxygen and hydrogen gas yield. Moreover, the third bipolar electrode produces carbon dioxide to provide a desirable anoxic condition (Zhang et al., 2005; Zhou et al., 2007; Ghafari et al., 2009).

Fully autotrophic nitrification and denitrification in a palm shell granular activated carbon twin-chamber upflow bio-electrochemical reactor (PSGAC-TCUBER) has neither been documented nor implemented in previous studies. Therefore, the aims of the present study were to investigate the effect of hydraulic retention time (HRT) and electric current (I) on nitrogen removal and to optimize the operating parameters of the reactor for SND. In this regard, the central composite design (CCD) is a Design-Expert software used in all of the experiments. The developed polynomial models analyzed the data obtained from different runs. The sufficiency of the models was investigated via analysis of variance (ANOVA) to provide the statistical findings and diagnostic checking tests of the models.

2. Methods

2.1. Experimental set-up

The PSGAC-TCUBERs were built from Plexiglas tubes as shown in the schematic diagram of the experimental apparatus in Fig. 1. The reactors provided the desired oxic and anoxic conditions at the anode and cathode zones, respectively, using electric intensity in the divided cell. The reactor was designed to consist of two columns joined together by a connection (Plexiglas tubes: 8 cm long and 6 cm in internal diameter) located 4 cm from the bottom. The adjoining parts separated the anode and cathode compartments from each other by a cation permeable membrane (CPM) (Nafion 117, Dupont Wilmington, DE) through a flange and O-ring type joint. Each column was 42 cm in height and had an inner diameter of 10 cm. A mesh plate stainless steel (type 316) as electrodes (hole size 1 × 1 mm and 0.1 cm thickness) was installed at the bottom of each column and screwed to the plates, 12 stainless steel rods (5 mm in diameter and 40 mm in height) to enhance the supply of electricity in the palm shell-GAC as anode and cathode zones. The copper wires, which were sealed with fast-drying epoxy coated with silicon sealant, were connected to the electrodes

through the current collectors. The wires were connected directly to a programmable DC power supply (IPS-3202, RS Components, England). The reactors were watertight at the bottom and top using O-ring flanges to prevent the transfer of atmospheric gases in anaerobic and anoxic zones. The 600 g PSGAC from Bravo Green Sdn. Bhd., Sarawak, Malaysia with a size of 2–4 mm was packed into both columns (anode and cathode) with a height of 22 cm as biocarrier and third electrode. The total volume of the reactor in each compartment was 3.5 L. The effluent ports were located 33 cm above the plate in the nitrification and denitrification chambers. The working volume for each column was 1.8 L, and the effective volume of 1 L was considered as anode and cathode zones. The void volume at the electrode zone was 1 L, and the ratio was 0.57. A number of sampling ports (SP) were installed along the opposite side of each column to inoculate the sludge on the PSGAC. Sampling ports played an important role in adjusting the pH between 7.6 and 8.0 via an automatic injection of acid (H_2SO_4 ; 1N) or alkaline (NaOH ; 1N) solution through a pH controller (Biostat B, B. Braun, Biotech International), which was connected to fermenter. A reverse cone-shaped funnel was used as a gas separator to transfer the produced gases through the outlet to the water seal. The controlling parameters (DO and pH) were monitored using DO meter (METTLER TOLEDO, O_2 -sensor, Switzerland) and pH meter (METTLER TOLEDO, pH-sensor, Switzerland) positioned at a height of 25 cm from the bottom of the reactors in both compartments.

2.2. Inoculated biomass and feeding

The mixed culture of acclimatized nitrifying bacteria and hydrogenotrophic denitrifying bacteria from prior research studies

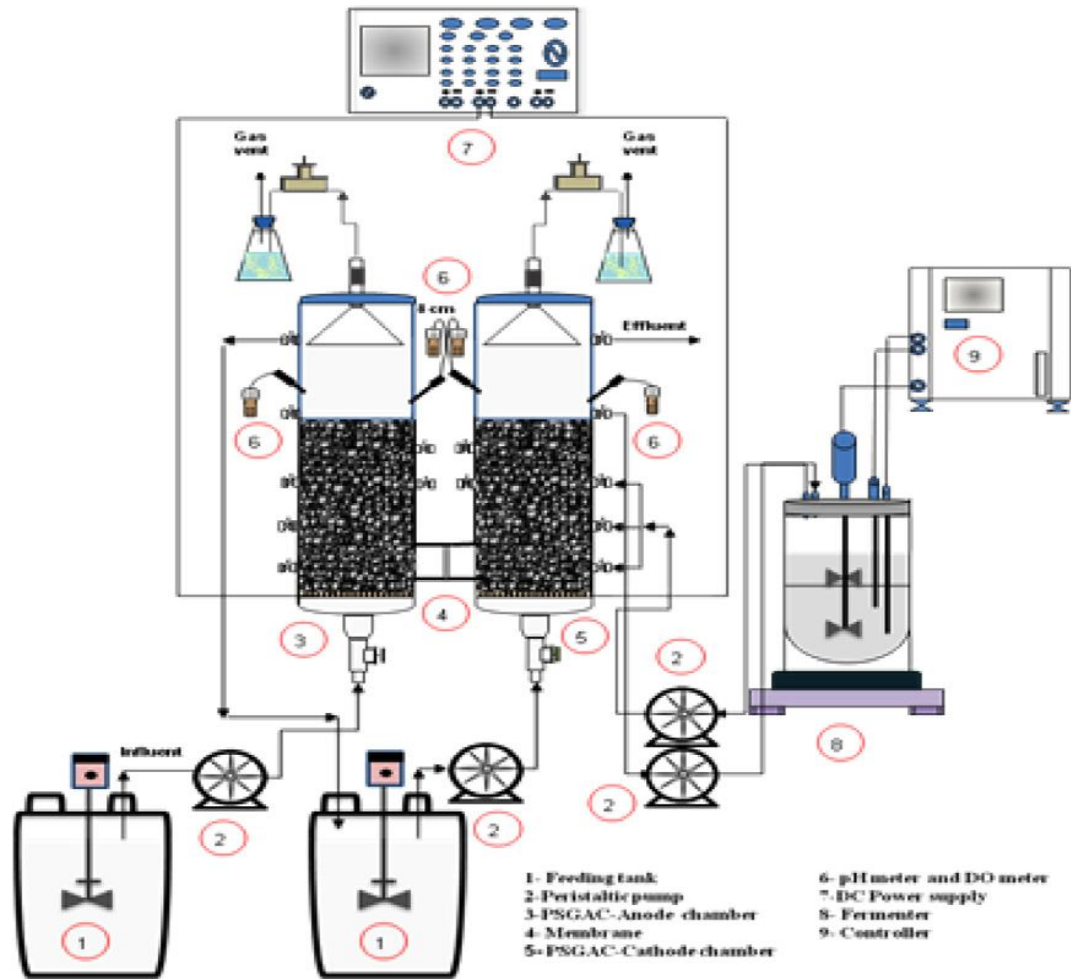


Fig. 1. Schematic of palm shell granular activated carbon twin chamber up-flow bio-electrochemical reactor (PSGAC-TCUBER).

inoculated into the TCUBERs after 20 days of re-acclimatization, to grow on the PSGAC. Before the injection of biomass inside the reactors, the following preparation of bio-carrier was performed.

The washed PSGAC was soaked several times in deionized water overnight. The deionized water was drained from the GAC, and then dried at 105 °C. The GAC was then saturated in 1% agar solution (Bacto™ Agar) at boiling temperature to produce a sticky surface. The cold PSGAC was transferred into BERs, and the re-acclimatized sludge, with initial mixed liquor suspended solids (MLSS) (3,000 mg L⁻¹), was pumped on the GAC columns through five sampling ports from the opposite sides of each compartment. The volumetric ratio of GAC to total reactor volume was 0.55. The growth and re-acclimatization of biomass on the GAC was maintained for 40 days by controlling the environmental and operational parameters: pH of 7.8 ± 0.2; T of 29 ± 1 °C; NH₄⁺-N of 200 mg L⁻¹

The BERs from the oxic part (anode compartment) was fed with synthetic wastewater according to Table 1. The anoxic part (cathode compartments) was fed with effluent from the first compartment (anode). To limit the interference and unsuitable effect of O₂ on the denitrification process, the nitrogen gas was sparged through an influent at a rate of 2 L min⁻¹ for 5 min to drive off the DO (<0.5 mg L⁻¹).

2.3. Analytical methods

The samples were analyzed immediately or were stored at cold temperature (4 °C) prior to the analysis. The samples for the determination of dissolved components, e.g., ammonium, nitrate, and nitrite concentration, were analyzed using an Advanced Compact Ion Chromatograph IC 861 (Metrohm Ltd., Herisau, Switzerland) and guard column. The effluents used to determine the cations and anions was prepared using ultra pure water (18.2 ls) containing pyridine-2,6-dicarbonsaure (0.117 g L⁻¹) and HNO₃ (0.11 mL L⁻¹) for cation components, and NaCO₃ (0.3392 g L⁻¹), NaHCO₃ (0.084 mL L⁻¹), and H₂SO₄ (0.1 M) for anion components, respectively. Samples were filtered before the analysis using a 0.2 μm filter. Temperature, pH, and DO were monitored continuously online. In addition, other experimental tests such as MLSS, mixed liquor volatile suspended solids (MLVSS), were determined using standard methods (APHA et al., 2005). The repetition of analysis was considered when an error higher than 5% was observed in the samples during the experiment. The ability of system for ammonium removal can be expressed in term of current efficiency (CE) according to Eq. (3) (Prosnansky et al., 2002; Ghafari et al., 2009).

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