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Carbon attrition during continuous electrolysis in carbon bed based three-phase three-dimensional electrode reactor: Treatment of recalcitrant chemical industry wastewater

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ABSTRACT

The performance of a carbon bed based three-dimensional electrode reactor (TDR) in terms of chemical oxygen demand (COD) removal from recalcitrant chemical industry wastewater was assessed. The pH and temperature changes in the TDR during electrolysis were correlated with COD removal efficiency. The carbon weight loss and particle size reduction due to erosion of carbon particles during electrolysis was also examined. Two cases of experiments were performed; 'case I' employed a high surface area Indcarb-60 GAC, whereas a low surface area carbon (GAC-10) was used for 'case II'. The other experimental variables are, initial COD concentration, hydraulic retention time (HRT) and the duration of electrolysis. The experimental results showed that TDR could remove COD efficiently ($49 \pm 7\%$). The apparent Faradic efficiency and specific electrical energy consumption were estimated to be 3.42% and 6.59 kW h kg^{-1} COD for case I and 0.78% and 28.65 kW h kg^{-1} COD for case II. Use of high surface area carbon in TDR is inferred to be beneficial. However, the GAC particles in TDR were found to undergo slow attrition during electrolysis. It is inferred that carbon attrition may prove to be a major setback for scale up attempts as it can lead to gradual loss in liquid holding capacity of carbon bed due to stratification and filling of voids in the carbon bed with carbon fine dust.

1. Introduction

High strength chemical industry wastewater requires high degree of treatment before being discharged into the water bodies. The conventional treatment incorporating chemical and biological treatments alone is not adequate. More recently, advanced oxidation processes such as electrochemical oxidation (EO) treatment are being increasingly viewed as capable of providing necessary treatment [1–4]. A variety of design options for setting up an electro oxidation reactor are available, viz., cell configuration (divided and undivided), electrode configuration (two-dimensional and three-dimensional) and flow-types (plug flow and continuously stirred tank type) [5]. Our group has been working towards the development of three-phase three-dimensional carbon bed electrochemical reactor (TDR) with the aim of treating recalcitrant effluents from chemical industries [6–10]. Thus, leachate from a toxic solid waste disposal facility could be treated effectively i.e., 60–64% COD removal with >80% mineralization efficiency using TDR in 6 h [7]. On the other hand, caprolactam wastewater underwent poorer degradation in TDR with only ~18% COD removal in 7 h [8]. While the above research clearly demonstrates that electro oxidation of recalcitrant effluents in TDR is feasible, there is need for optimizing the operational parameters (pH, flow rate etc.). Moreover, the electrochemical degradation of wastewater using such TDR in continuous mode is not comprehensively investigated, particularly with reference to stability of carbon surface on long term use.

In this study, continuous electrolysis of chemical industry wastewater in the TDR was investigated. The performance of the reactor for consistent removal of pollutant (COD) was aimed at, while the other related issues viz., stability of carbon particles, changes in pH, and temperature were also investigated.

2. Material and methods

2.1. Effluent

The segregated high strength chemical industries wastewater used in this study was obtained from a common effluent treatment plant (CETP) site in Gujarat (India). The dark brown effluent with pH 8.1 contained high COD in the range 5–10 g L^{-1}, 1.1–2.4 g L^{-1}

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TKN (Total Kjeldahl Nitrogen), 0.35–0.9 g L^{-1} ammonia nitrogen (NH_4^+-N) and 17–22 g L^{-1} chloride. The wastewater used for the present study may be regarded as non-biodegradable high strength wastewater (BOD/COD ≤ 0.2).

2.2. Granular activated carbon

Two different types of granular activated carbon (GAC) were used, Indcarb-60 (IC-60) with particle size ranges from 2 to 3 mm, 1100 m^2 g^{-1} surface area and GAC-10 with particle size >4 mm, surface area 600 m^2 g^{-1}. The GAC was purchased from Industrial Carbons Pvt. Ltd., Baroda, India and it was used in the three dimensional carbon bed electrochemical reactors.

2.3. Reactor setup and experimental procedure

Continuous carbon bed electrolysis of high strength chemical industry wastewater was carried out using three dimensional carbon bed electrochemical reactors described elsewhere [7,8]. The schematic of the TDR is given in Fig. 1. The TDR was a rectangular 2.6 L tank (18 cm × 12 cm × 12 cm) fabricated using the Perspex® sheet of 6 mm thickness. In this reactor, two stainless steel plates (10 cm × 14 cm) were used as cathode and situated 12 cm apart on opposite side walls of the reactor. In the middle of the reactor a 12 mm carbon plate was used as anode in between SS cathodes. This carbon anode was perforated by drilling 2 mm diameter holes to allow free flow of liquid across anode. Approximately 1 kg of washed and dried GAC was packed into the space between the cathode and anode to form a three dimensional electrode (GAC bed height, 10 cm) and air was bubbled from the bottom of the reactor through an air distribution manifold. Current/potential was applied using a regulated direct current power supply (Aplab, LD3210). Before electrolysis experiment, the TDR bed was pre-saturated by circulating the raw effluent (10 L, pH 8.1) that was fortified with NaCl (2 g L^{-1}) through the TDR bed at a rate equal to 0.1 L min^{-1} by using peristaltic pump (Watson-Marlow, 323 DU).

Two types of experiments in continuous carbon bed electrolysis of wastewater were performed: case I – it was carried out by using indcarb-60, HRT of 2 h for 5 days with raw effluent (COD, 10,358 mg L^{-1}) and case II employs GAC-10, HRT of 4 h for 10 days and raw effluent having COD, 5200 mg L^{-1} was used. Continuous reaction was performed at 3 A (constant current) and 5.5–7.9 V. Aliquots were taken from the outlet of the reactor at pre-selected time intervals and analyzed for COD reduction. GAC weight loss study was also performed. Experiments in continuous operation with the different current viz., 1, 2, and 3 A were carried out using brine electrolyte. Fresh GAC-10 (1 kg) was packed in TDR and 50 L of sodium chloride solution (18 g L^{-1}) passed through in each experiment with constant HRT of 4 h. GAC after electrolysis was washed, dried overnight at 110 °C and sieved (ASTM 5). GAC particle size <4 mm was collected and weighed to assess the weight loss gravimetrically. The wash water (10 L) was filtered through glass fiber (1.2 μm) and residue collected was dried overnight at 110 °C and weighed.

The apparent Faradic efficiency (η_F) of COD removal was calculated using following equation [7,8,10].

$$\eta_F = \frac{\Delta \mathrm{COD} \times V \times F}{8 \times I \times \Delta t} \quad (1)$$

where ΔCOD is the net COD removed (g L^{-1}) after a treatment time t (h), V is the volume of treated effluent (L), F is Faraday's constant (96,487 C equivalent^{-1}), 8 is the equivalent weight of oxygen, I is the applied current (A) and Δt is the treatment duration (s).

Specific energy consumption (E_{sp}), the electric energy in kilowatt hours required to degrade a kilogram of a pollutant in wastewater, was calculated using the following equation for continuous mode of operation [7,8,10].

$$E_{SP} = \frac{P \times t \times 10^6}{V(C_0 - C_t)} \quad (2)$$

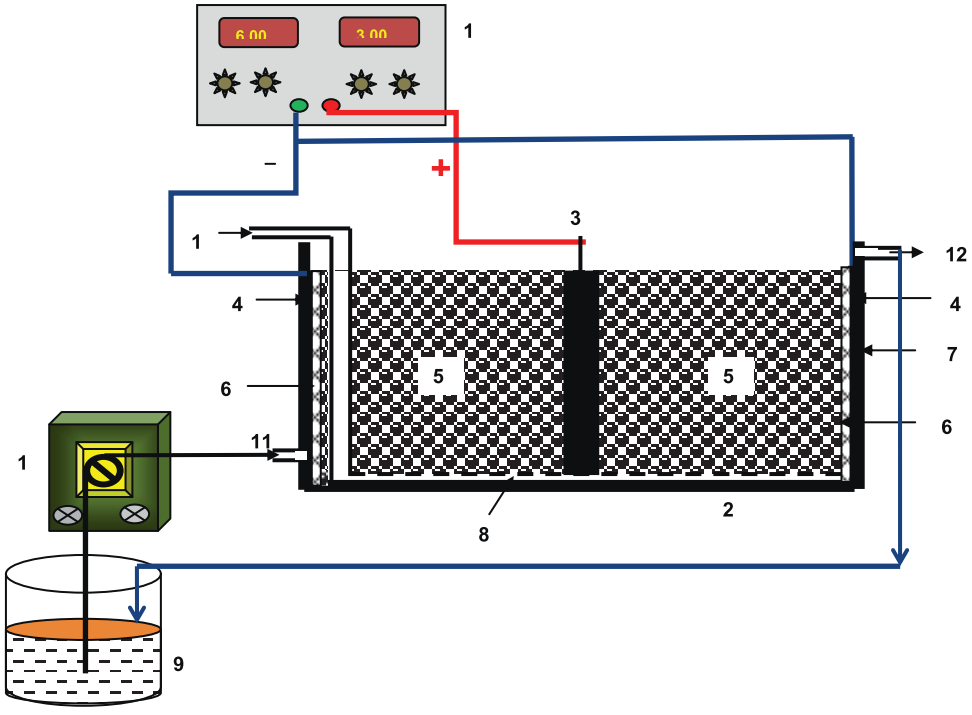

Fig. 1. Schematics of carbon bed based three-dimensional electrode reactor (TDR).