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Short Communication

Efficient treatment of domestic wastewater by electrochemical oxidation process using bored doped diamond anode

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ABSTRACT

Domestic wastewaters (DWW) contain varied and high amounts of organic matter which are difficult to oxidize biologically or chemically. The discharge of such effluents is undesirable and can cause excessive oxygen demand in the receiving water. In order to produce an effluent suitable for stream discharge, electrochemical techniques have been explored at the laboratory pilot scale for organic compound removal from DWW. Electrolysis of DWW was carried out using rectangular electrolytic cell containing two circular electrodes (anode and cathode). A preliminary voltametric study was carried out using different anode materials (Ti/BDD, Ti/IrO_2 and Ti/IrO_2–RuO_2). The Ti/BDD electrode was found to be the most effective electrode for DWW treatment. Electrochemical oxidation process using Ti/BDD electrode at the anode and a-C at the cathode with a current density imposed of 26.53 mA/cm^2 ensures the removal efficiencies of 78.2%, 70% and 89.5% of COD, TOC and color, respectively. DWW could be oxidized by both direct anodic electrochemical oxidation (by means of $OH^{\cdot}$) and indirect electrochemical oxidation via mediators, such as hydrogen peroxide generated by cathodic reduction. Both actions (direct and indirect effects) lead to the formation of powerful oxidizing agents capable of oxidizing refractory organic compounds.

1. Introduction

Domestic wastewater (DWW) generated from isolated communities and industries are complex mixture of organics, inorganic and microbial pollutants [1]. Their exposure and accumulation in the aquatic environment lead to adverse effects towards human life and cause eutrophication of surface waters and transmission of waterborne diseases [2,3]. Therefore, the major concern is to treat the wastewater before discharge into the aquatic environment. The stricter restrictions imposed by new legislations have caused extensive effort to focus on the development of alternative and effective treatment processes in order to remove organic pollutants from decentralized wastewaters.

Nowadays, electrochemical advanced oxidation processes have been proposed and identified as an attractive option for wastewaters treatment [4–6]. Electrochemical advanced oxidation processes are environmentally friendly technologies capable of producing hydroxyl radicals ($E^{\cdot}(OH^{\cdot}/H_2O)$ = 2.80 V vs. SHE). Hydroxyl radicals are very powerful oxidants capable of oxidizing a wide range of organic compounds [4,5]. The Electrooxidation (EO) process has been found to be a promising environmental remediation technology to remove organic pollutants [7–9]. The interest of using EO is based on its capability of reacting on pollutants by using both direct and indirect effect of electrical current [9–11]. Direct oxidation may be achieved through mineralization with hydroxyl radicals ($OH^{\cdot}$) produced at the electrode surface by dimensionally stable anodes (DSA) having high oxygen overvoltage, such as IrO_2, PbO_2 and boron doped diamond (BDD), among others [12–14]. In fact, $OH^{\cdot}$ radicals are exclusively generated on the anode electrodes from the oxidation of water (Eq. (1)) and organic compounds can be completely transformed or degraded by reacting with adsorbed $OH^{\cdot}$ radicals (Eq. (2)) [15–17]. As reported by previous research works [17,18], the electrochemical oxidation process using BDD as anode is highly potent to provide more rapid destruction of pollutants and total mineralization of the organic loading with high current efficiencies. Compared to the other DSA, BDD has received a great attention due to its inert surface with low adsorption properties, its remarkable corrosion stability even in acidic media and its high oxygen evolution over-potential [10,19].

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$$M + H_2O \rightarrow M(\cdot OH) + H^+ + e^-) \quad (1)$$

$$M(\cdot OH) + \text{organics} \rightarrow M + \text{oxidation products} \quad (2)$$

Furthermore, indirect oxidation can be achieved through electrochemical generation of a mediator in solution such as H_2O_2 [20,21]. Hydrogen peroxide (H_2O_2) (1.77 V) electrochemically generated at the cathode is an environmentally metastable molecule with high disinfecting and oxidizing properties able to convert toxic organics to a less harmful products [20,22].

In this context, it could be interesting to explore at the laboratory scale the electrochemical oxidation process in order to remove organic pollutants from domestic wastewater (DWW). The DWW was an effluent provided from a tank (septic tank) and collected after bio-filtration treatment of wastewater from isolated residences. The main objective of this study is to develop an electrochemical oxidation process using boron doped diamond at the anode and to evaluate its performance in treating domestic wastewaters.

2. Materials and methods

2.1. Sampling and characterization of DWW

The domestic wastewaters (DWW) used throughout this study was an effluent provided from Riviere du Loup community domestic wastewater treatment plant (WTP, Riviere du Loup, Quebec, Canada). It is a conventional WTP having a primary treatment (septic tank) of wastewater providing from isolated residences followed by a bio-filtration process. The effluents were sampled at the outlet of the bio-filtration unit. Samples were collected and stored in polypropylene bottles and kept at 4 °C until use. The treated-effluent had an initial content of COD of 79.17 ± 4 mg/L, TOC of 19.9 ± 0.8 mg/L, color of 100 ± 4 TCU, turbidity of 11.4 ± 0.3 NTU, pH of 7 ± 0.03 and conductivity of 941.7 ± 10 μs/cm.

2.2. Electrolytic cell

The electrolytic cell used was made of PVC material with a dimension of 17 cm (depth) × 5 cm (width) × 15 cm (length). The electrochemical cell was comprised of one anode and one cathode electrodes in the form of expanded metal, each having a solid surface area of 65 cm^2 and a void surface area of 45 cm^2. The circular anode electrode (12 cm of diameter × 0.1 cm thick) was either made of titanium coated with boron doped diamond (Ti/BDD), whereas the circular cathode electrode was made of titanium coated with amorphous carbon (Ti/a-C) (12 cm of diameter × 0.1 cm thick). The magnetron-sputtering technique was used to coat deployed titanium grid substrates with nanostructured amorphous carbon (a-C) films. The inter-electrode gap was 1.0 cm in the electrolytic cell. The electrodes were vertically installed on a perforated Plexiglas plate at 2 cm from the bottom of the cell.

2.3. Experimental setup

The tests were carried out in a closed loop as depicted schematically in Fig. 1. A 2.0 L of PVC tank (1), a peristaltic recycling pump (2) and the electrolytic cell (3) constitute the loop. All the essays were conducted in batch recirculation mode with a flow of effluent entering the bottom of the cell. A recycle flow rate of 120 mL/min was maintained using a peristaltic recycling pump (Master flex, Model 77200-50, USA). The electrochemical cell was operated under galvanostatic conditions, with a current intensity (from 0.5 A to 3 A) imposed during 120 min of treatment period. Current intensity was imposed by means of DC power supply Xantrex XFR 40-70 (Aca Tmetrix, Mississauga, ON, Canada) with a maximum current rating of 70 A at an open circuit potential of 40 V. All experiments were carried out at room temperature (25 ± 0.1 °C). For all tests, a total volume of 1.0 L was used.

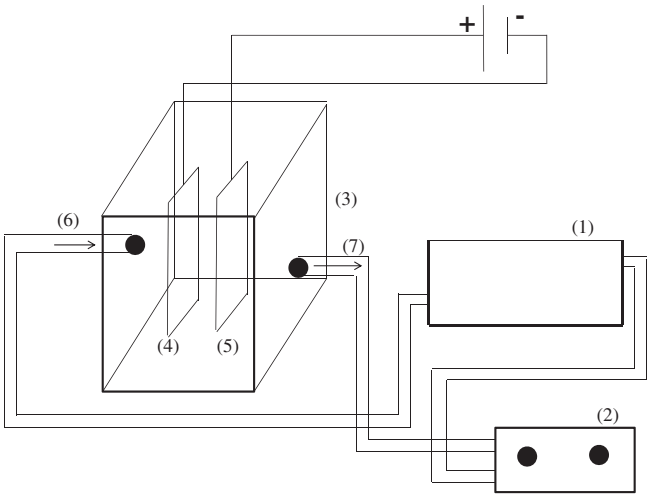

Fig. 1. Schematic diagram of the experimental setup: (1) PVC tank, (2) peristaltic recycling pump, (3) electrolytic reactor, (4) anode, (5) cathode, (6) in let, (7) out let.

2.4. Experimental procedure

A preliminary voltammetric study was performed with a three-electrode Voltalab 80 system (Radiometer analytical) equipped with a rotating working electrode (CVT 101, Radiometer analytical). It included a potentiostat/galvanostat (Tacussel 20V/1A PGP 201). The reference electrode was a saturated calomel (Hg/Hg_2Cl_2/KCl) electrode (Tacussel XM 110), while the auxiliary electrode was platinum (Taccusel Pt XM 140). The materials used as working electrode were titanium coated with bored doped diamond (Ti/BDD, 11 mm diameter), titanium coated with iridium oxide (Ti/IrO_2, 11 mm diameter) and titanium coated with iridium-ruthenium dioxide (Ti/IrO_2–RuO_2, 11 mm diameter). Linear voltammetric measurements were conducted in a 0.1 M Na_2SO_4 electrolyte solution to identify the best electrodes. Subsequently, the treatment of DWW using electrochemical oxidation process was operated under different operating parameters such as electrodes materials (Ti/BDD; and Ti/IrO_2–RuO_2) and current density (from 4.42 mA/cm^2 to 26.5 mA/cm^2) in order to determine the best conditions (reduce cost and increase effectiveness). Experiments were carried out over 120 min of electrolysis times. Owing to the high conductivity, the treatment of DWW by electrochemical oxidation process was performed without adding Na_2SO_4. During these assays, the effectiveness of the electrochemical oxidation process was evaluated by simultaneously measuring the residual concentration of COD, TOC and color. Energy consumption of each test was also determined. To verify the effectiveness and the reproducibility of the electrochemical oxidation process, the measurement of all parameters were repeated in triplicate.

2.5. Analytical technique

The pH was determined using a pH-meter (Fisher Acumet model 915) equipped with a double junction Cole-Palmer electrode with Ag/AgCl reference cell. A conductivity meter (Oakton Model 510) was used to determine the ionic conductivity of the effluent. Turbidity (in nephelometric units, NTU) was measured using a turbidimeter Hach 2100 AN. Chemical oxygen demand (COD)