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Electro oxidation of dye effluent in a tubular electrochemical reactor using TiO_2/RuO_2 anode

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ABSTRACT

The aim of the present investigation is to treat Evans blue dye effluent using tubular electrochemical reactor. The electrode is made of stainless steel and oxide coated expanded titanium as cathode and anode respectively. The cylindrical mesh type anode is fitted in the tubular electrochemical reactor. The effect of operating parameters such as current density; initial dye concentration; initial solution pH, recirculation flow rate and supporting electrolyte concentration has been studied on COD reduction. It can be observed from the present investigation that the electrode $Ti/RuO_{0.3}Ti_{0.7}O_2$ effectively degrade the Evans blue dye effluent and the kinetics of Evans blue dye degradation follows the pseudo first order kinetics. Further, dispersion model has also been developed to determine the exit concentration of the effluent. The model simulation is compared with experimental value and it is observed that the model matches well with correlation coefficient of 0.98.

1. Introduction

The industrial growth and urbanization has resulted in an increase the quantity and complexity of wastewater to the environment. Textile industries are one of the most polluting industries in terms of the volume and complexity of its effluent discharge. The dyeing and finishing operations in textile industries generate a major wastewater. These processes require huge volume of water and as a result, generating large volume of wastewater which have high COD, BOD and strong color. These effluents are liable to cause environmental issues if they are discharged without proper treatment. Hence the treatment of the effluent is mandatory before letting out to the environment [1].

Various conventional techniques such as flocculation, coagulation, membrane-based nanofiltration, reverse osmosis, adsorption process and activated sludge processes are widely used for the wastewater treatment. However, these processes are inadequate in treating effluents which are recalcitrant nature. These difficulties are well handled using electrochemical method which has the unique advantages such as versatility, energy efficiency, automation and cost effectiveness [2].

Electrochemical methods have been successfully applied in treatment of several industrial wastewaters [3–7]. In the electrochemical method of treating effluents, the electrode material and reactor design play an important role. Dimensionally stable anode (DSA) such as SnO_2, PbO_2, IrO_2, boron doped diamond (BDD), Ti/Pt, TiO_2 *etc.*, has been found wide application in the electrochemical treatment of effluents. In any electrochemical process, electrode material and electrochemical reactor design is important. The electrochemical reactors varying from small scale batch reactor to large size flow cells. The choice of batch or continuous operation for industrial application depends on the nature and toxicity of the effluent, simplicity of the process required, and the process economics [8]. The choice of electrode material is of fundamental importance from electrochemical point of view. Boron doped diamond and dimensionally stable anode (titanium substrate is covered by metallic oxides such as TiO_2, IrO_2, RuO_2, SnO_2) materials were widely studied for application in waste treatment systems [9–18].

Various electrochemical reactors ranging from batch to continuous operations have been used for synthesis of chemicals and waste water treatment application using different electrodes. Korbahti and Tanyoalc [19] studied phenolic wastewater treatment in a continuous tubular reactor using stainless steel cathode and carbon anode. The authors reported that 95% of non-passivating polymer was produced without producing any hazardous product. Raghu and Ahmed Basha [20] studied Procion Black 5B dye treatment using stainless steel as a cathode and Ti/RuO_2 as anode in a cylindrical flow electrochemical reactor. The author observed 100%, 74% of color and COD removal respectively using this reactor.

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Muthukumar et al. [21], studied textile dye wastewater treatment using RuO_2 coated titanium as anode and titanium as cathode in a bipolar disc stack reactor. The authors reported that higher COD removal was achieved at higher flow rate and lower reservoir hold-up. Ahmed Basha et al. [22], studied simultaneous removal of copper and phthalocyanine from a real acidic effluent of copper phthalocyanine dye manufacturing plant using a bipolar disc electrochemical reactor. Around 91.1% COD removal and 90.1% copper recovery was observed in a bipolar disc stack reactor using RuO_2 and IrO_2 coated on titanium as anode and titanium as cathode. Bhadrinarayana et al. [23], studied electrochemical oxidation of cyanide and cadmium removal using graphite electrode in the bipolar disc stack reactor. The author observed 99% cyanide destruction and 98% removal of cadmium in the plating rinse water. Chellammal et al. [24], studied the process wastewater coming out from the copper phthalocyanine manufacturing plant in a galvanostatic batch reactor using stainless steel as cathode, DSA and graphite as anodes and optimized conditions for the maximum copper recovery of 98% and COD removal efficiency of 87.3%. Electro oxidation of distillery industry wastewater using a flow electro chemical reactor in which graphite particles with titanium sponge as the voluminous anode and Ti/RuO_2 as a cathode are studied by Puja-areetham et al. [25]. The authors reported that the maximum COD removal was achieved in acidic pH condition. Vlyssides et al. [26] studied laboratory scale pilot plant using Pt/TiO_2 anode in the presence of sodium chloride as supporting electrolyte. The author observed 89% of COD removal for an influent having COD concentration of 72,000 mg L^{-1}. Electrochemical degradation of Acid Red 73 in aqueous solution using Ti/SnO_2-Sb-CNT electrode is studied by Xu et al. [27]. The author observed 95.7% dye removal and the kinetics follows the pseudo first order.

Various literatures have been reported the degradation of the pollutant using dimensionally stable anode. But no attempts were found for the modeling of the reactor used in the waste water treatment. Hence the objective of the present investigation is to carry out experimental as well as modeling of the reactor for waste water treatment using electrochemical technique. The study focuses on kinetics of the degradation of Evans blue dye effluent in a tubular electrochemical reactor using Dimensionally Stable Anode (DSA) ($Ti/Ru_{0.3}Ti_{0.7}O_2$) and stainless steel as cathode under recirculation mode. Further the influence of operating parameters on COD reduction has also been investigated.

2. Materials and method

All the reagents used for the present study is AR grade. Evans blue dye obtained from Sigma–Aldrich is used without further purification. The synthetic effluent was prepared from Evans blue dye dissolved in water. H_2SO_4 and NaOH were used to alter the pH of the solution. NaCl is used as a supporting electrolyte to increase the conductivity of the solution.

2.1. Tubular electrochemical reactor

The electrochemical reactor flow cell model used for the study is shown in Fig. 1. Electrochemical experiments were carried out using a stainless steel cathode and a $Ti_{0.7}Ru_{0.3}O_2$-coated titanium substrate as an insoluble anode ($Ti/Ti_{0.7}Ru_{0.3}O_2$). That means the coating content is 70% TiO_2/30% RuO_2 by weight percentage. The cathode was made of stainless steel with a diameter of 7 cm and a height of 110 cm. The thickness of the tube is 2 mm. At the top edge of the tube, provisions were made for power supply, while the mesh type anode having 60% perforation of tubular shape with dimension of 5 cm diameter and height of 100 cm which resulted in an effective anode area of 628.3 cm^2 (1570.8 cm^2 × 0.4) The anode was

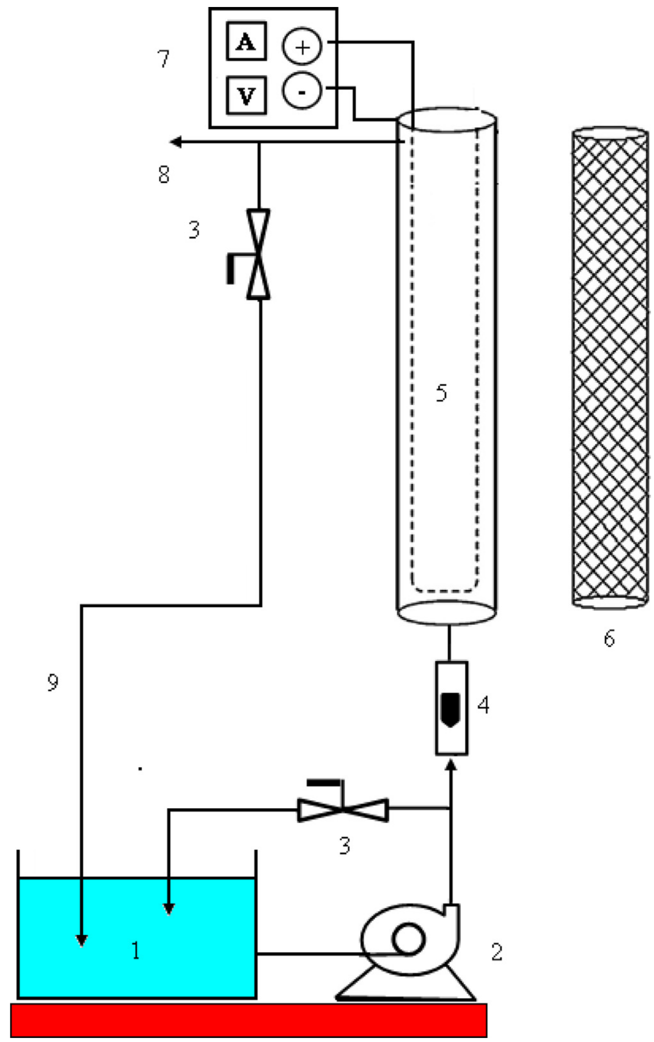

Fig. 1. Schematic diagram of tubular electrochemical reactor with recirculation: (1) reservoir, (2) centrifugal pump, (3) control valve, (4) rotameter, (5) tubular reactor, (6) cylindrical mesh anode (7). DC power supply (8) reactor outlet (9) recirculation mode of operation.

inserted inside the cathode, and was sealed with the end frames. Electrodes were connected to 100 A and 0–50 V DC regulated power supply. Reservoir, pump and flow meter were connected using silicone rubber tubes. In the present investigation a batch recycling operation was performed to treat the dye effluent.

2.2. Experimental procedure

Synthetically prepared Evans blue dye effluent was taken in the reservoir with different concentration. The effluent was passed through the tubular electrochemical reactor. The supporting electrolyte concentration is varied from 1 g L^{-1} to 3 g L^{-1} and pH is varied as 4, 7 and 9. Three different flow rates (25, 50, 75 L h^{-1}) were measured by adjusting the rotameter. At each current density, experiments were conducted for different flow rates. Reactor is cleaned by washing with water before every run. All experiments were carried out for 90 min in batch recirculation mode under galavanastatic conditions. Each experimental runs, samples were collected and subjected to COD analysis.

The COD reduction is estimated as:

$$(1 - X) = \left(\frac{C}{C_i}\right) = \frac{\text{Final COD of the effluent}}{\text{Intial COD of the effluent}} \quad (1)$$