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Development of Box Behnken design for treatment of terephthalic acid wastewater by electrocoagulation process: Optimization of process and analysis of sludge

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ABSTRACT

Present research work deals with the treatment of terephthalic acid (TPA) wastewater by electrocoagulation (EC) process. TPA is counted in the major toxic pollutants of purified terephthalic acid (PTA) wastewater and contributes high COD. Box Behnken Design (BBD) in Response Surface Methodology (RSM) tool of Design Expert Software (8.0.7.1, 2010, Stat-Ease Inc., Minneapolis) has been developed for this study. Effect of various operating parameters viz. pH: (7 to 11), current density (A/m^2): (38.40 to 105), electrolysis time (min): (15.0 to 60.0) and NaCl dosages (g/L): (0.50 to 1.25) on percentage removal of TPA, chemical oxygen demand (COD) and energy consumption (kW h/kg $COD_{removed}$) are presented. Maximum percentage removal of TPA: 89.75, 73.86; COD: 83.88, 67.63 and minimum energy consumption (kW h/kg $COD_{removed}$): 57.25, 70.84 are found at optimum operating conditions using Al and Fe electrodes respectively. Properties of sludge were analyzed by point of zero charge, settling, XRD, SEM/EDX, FTIR and TGA/DTA. Process operating costs ($) (Al: 8.59, Fe: 9.82) are determined based on electrical energy, electrode consumption and electrode maintenance cost for per kg of COD removal at optimum operating conditions.

1. Introduction

Purified terephthalic acid (PTA) wastewater comprises terephthalic acid (TPA), benzoic acid (BA) and para-toulic acid (*p*-TA) as main aromatic compounds contributing ~75% COD [1,2]. TPA is the para form of phthalic acid (PA) (1,4,benzene di-carboxylic acid). It is also used in various applications viz. synthesis of polyethylene terephthalate (PET) and poly-butyl terephthalate (PBT). These compounds are used in manufacturing of carbonated/soft drinks bottles, clamshell, plastics, pesticides, polyesters, textiles, fibers, adhesive, plasticizer, polyester films, coating materials, dyes, packing materials and molded resins etc. [3–5]. TPA is also used in manufacturing of poultry feed additives, medicines, synthetic perfumes, bio-plastics and other chemical compounds [6].

Although, TPA has various applications but due to its toxic nature intake in higher amount is dangerous for human body and microorganisms. It shows non-toxic behavior at concentration below 15 mg/L at 10 °C and intake above this may cause bladder cancer, bladder stones, and impairment of testicular functions. It may also affect the reproductive and development system of human being, can cause gene mutation and renal disfunction in animals [7–10]. It mildly irritates eyes, skin, and respiratory system [11]. It also shows acute toxicity, chronic toxicity, sub-acute toxicity, and molecular toxicity [12,13]. Due to its toxic behavior, USEPA added TPA in the list of priority pollutants [14].

About 3–4 m^3 of wastewater is generated during production of one tonne of TPA with very high COD (5–20 g/L) [11,15]. In India, pollution-regulating agencies like Central Pollution Control Board (CPCB) and Ministry of Environment and Forests (MOEF) have stipulated maximum permissible limit of COD as <250 mg/L for discharge of petrochemical wastewater into surface water [16]. Since last few decades, TPA wastewater has been treated by various processes viz. biological—aerobic and anaerobic, physiochemical, advance oxidation processes (AOPs), thermo-chemical precipitation, electrochemical treatment-electrocoagulation [17–28].

Abbreviations: ANOVA, analysis of variance; BA, benzoic acid; BBD, Box Behnken design; CD, current density; COD, chemical oxygen demand; DTA/TGA, differential thermal analysis/thermo gravimetric analysis; FTIR, Fourier transform infrared spectroscopy; PA, phthalic acid; PRESS, predicted residuals error sum of squares; *p*-TA, para-toulic acid; PTA, purified terephthalic acid; PZC, point of zero charge; RSM, response surface methodology; SEM/EDX, scan electron microscopy/energy dispersive index; TPA, terephthalic acid; XRD, X-ray diffraction.

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Electrocoagulation (EC) is an emerging technology used for treatment of various industries wastewater owing to its in-situ generation of coagulating agents by electro-oxidation of the anodes. EC process has added advantages over other conventional methods requiring simpler and low cost equipment, small footprint area, and short reaction time. It does not require any addition of coagulant or flocculent, chemical storage and efficiently removes smallest colloidal particles [29–33].

In recent years, Response Surface Methodology (RSM) in Design Expert Software has been used for optimization of various processes and specifically for wastewater treatment of phenol, chicken processing, rice mill, pulp and paper, color, polysaccharide, etc. [34–39]. Box Behnken Design (BBD) in RSM is an important design tool used for optimization of processes. BBD provides comprehensive conclusions and detailed information even for smaller number of experiments and interactive effects of operating parameters on all responses [40].

Treatment of TPA wastewater by EC process is an important step in this direction. A few studies have reported treatment of TPA wastewater by EC process [27,28] and these studies lack on optimization of process parameters, operating cost and analysis of generated sludge.

Accordingly, this study was under taken for treatment of TPA wastewater by EC process using Al and Fe electrodes to quantify the removal efficiencies of TPA, COD and energy consumption. Further, the optimization of the process using BBD in RSM tool of Design Expert Software (8.0.7.1, 2010, Stat-Ease Inc., Minneapolis) has been carried out. The experimental and model predicted data have been analysed. The sludge obtained at optimum operating conditions using Al and Fe electrodes has been analysed by settling, PZC, XRD, FTIR, TGA/DTA and SEM/EDX. The operating cost of the process based on the energy consumption, electrode consumption and electrode maintenance cost is also reported.

2. Materials and methods

2.1. Chemicals and wastewater sample preparation

2.1.1. Chemicals

Analytical reagent (AR) grade chemicals were used during entire study. TPA was supplied by Himedia Lab. Pvt. Ltd., Mumbai, India. All other chemicals viz. acetic acid, isopropyl alcohol, mercury(II) sulfate, methanol, potassium dichromate, sodium hydroxide, sodium chloride and sulfuric acid were supplied by Ranbaxy Chemical Ltd., New Delhi, India.

2.1.2. Wastewater sample preparation

Standard solution of TPA (1000 mg/L) was prepared at laboratory scale. Since TPA shows poor solubility at neutral or acidic pH during the preparation of standard solution of TPA wastewater, the pH of the solution was increased by addition of NaOH equivalent to the total molality of H^+ ions resulting in increased solubility of TPA. Water used for entire experimental study was purified by using Millipore milliQ system. All samples were preserved at 4 °C to reduce unwanted microorganism growth and biodegradation. Initial concentration of TPA (400 mg/L) was used in accordance to previous studies [21,27]. It was estimated that this wastewater had initial COD of 567 mg/L.

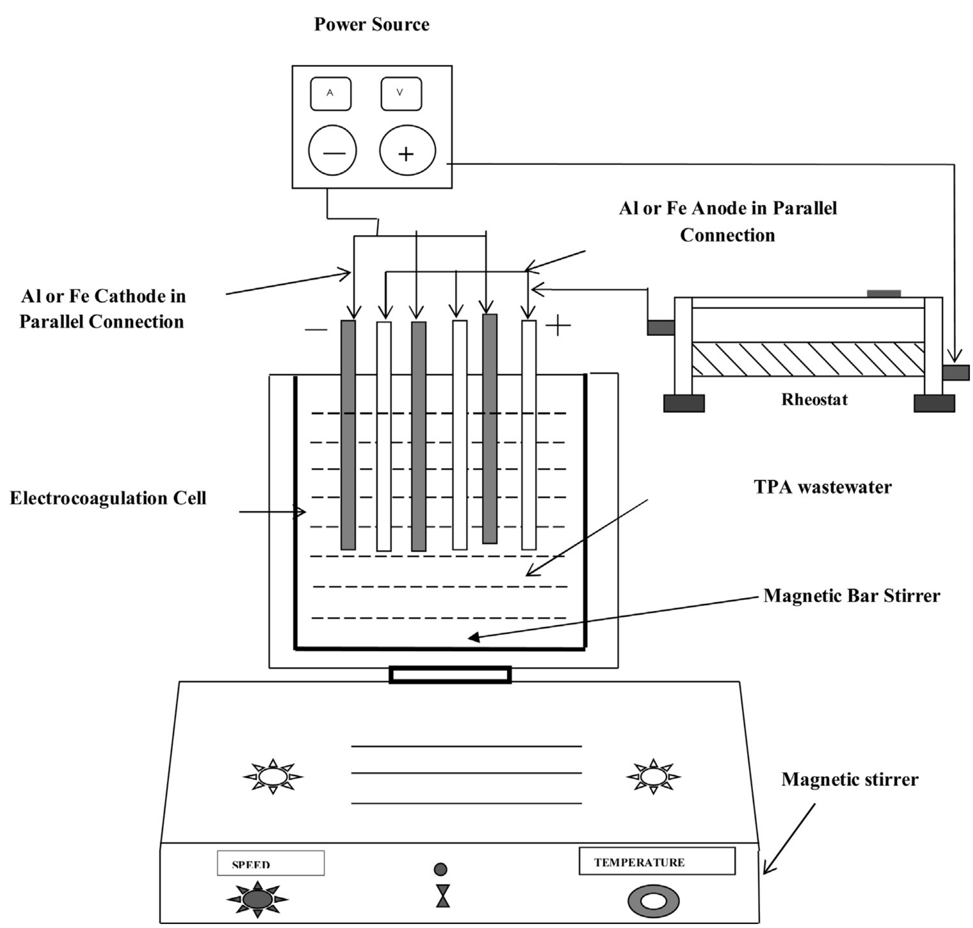

Fig. 1. Schematic view of the experimental system.