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An investigation on flow dynamics in the electrochemical reactor for the treatment of pharmaceutical wastewater

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ABSTRACT

In this study, the flow dynamic behavior of cylindrical electrochemical reactor (CER) has been investigated for the treatment of pharmaceutical effluent. The residence time distribution is considered as a tool to investigate the flow dynamic behavior of the electrolyte within the reactor. The reactor is operated at fixed current density of 2.5 A/dm^2 with lead oxide/ titanium as electrode by varying flow rates such as 50, 60, 70, 80 and 90 L/h. Impacts of various flow rates on flow dynamics were examined. The outcomes of this study demonstrate the presence of a dead volume and short circuiting in the reactor were reduced for the optimum flow rate of 70 L/h in the reactor. The potential of the CER was experimentally validated by analyzing the chemical oxygen demand (COD) removal efficiency, color, total dissolved solids, suspended solids and odor emanating from the effluent. Findings of this study reveal that maximum COD reduction of about 85.71% with minimum energy consumption of 63.82 kW h/kg COD at a flow rate of 70 L/h, which has good mixing and less backmixing condition inside the reactor. The experimental findings prove that the CER with a cylindrical mesh electrode can be used for the treatment of pharmaceutical effluents and is capable of achieving the quality of pharmaceutical effluents treated wastewater to the reuse standard prescribed for pharmaceutical industries.

Keywords: Electrochemical reactor; Flow dynamics; Residence time distribution; Pharmaceutical effluent; Electro-oxidation

1. Introduction

The pharmaceutical industry is overwhelmed with high-value life-saving drugs, low volume multiproduct plants on one hand that are mostly batch operations wherein the effluent is mixed and treated [1,2]. Water is a prominent raw material in pharmaceutical and chemical manufacturing operations; consistent and superior quality water supplies are needed for various operations, such as production, processing and cooling. The broad categories of water that need treatments as part of water management are potable water, process water, feed water for utilities, water recycling, wastewater, water coming from by-product treatment, water used for odor treatment, water from desalination and water for irrigation [3]. Process water quality management is of utmost important in pharmaceuticals manufacturing and is also an uncompromised requirement for the sterilization of medical devices in other healthcare applications including water for injection. Process wastewater is a general term used to define wastewater in any industry that is a by-product of its various processes. Process wastewaters thus mean any water that comes in contact with the raw materials, products, intermediates, by-products or waste products during manufacturing or processing, which are mainly handled in different unit operations or processes [4,5]. Modern pharmaceutical industries employ numerous wastewater treatment and disposal methods. Wastewaters generated from these industries vary not only in composition, but also in quantity.

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Plant geographical location also brings in a variable related to the quality of available water [6,7].

Electrochemical technology provides an ideal solution to address the environmental problems caused by these industries. In industries various electrochemical reactors are used for the waste water treatment applications such as tank cells, plate and frame filter press cells, cylindrical and tubular electrochemical reactors, rotating cells, three-dimensional electrode reactor systems like fluidized bed, packed bed cell or porous carbon packing cells. The materials (electrode) greatly used for waste water treatment in electrochemical industry are dimensionally stable anodes (DSAs) made of titanium (Ti) substrate coated with thin layer of noble metal oxides. The electro-oxidation of different kinds of wastewater had been investigated by numerous researchers using different types of the electrochemical reactors [8]. Of the other electrochemical reactors, the most common type of electrochemical reactor which is extensively used is cylindrical reactor and is widely studied by the researchers for the wastewater treatment and modeling [9]. The fluid flow inside the cylindrical electrochemical reactor (CER) is often assumed as dispersed plug flow of fluid. CER has higher mass transfer coefficients even at low axial flow rates thereby showing improvement in the pollutant removal rate with lower energy consumption.

In the electrochemical reactor system, effluents containing oxidizable species as pollutants can be considered as the once amiable for effective treatment by electro-oxidation process [10,11]. Electron is the main reagent used in the electrochemical process, which avoids the addition of other reagents [12,13]. During the process, the pollutants are breaked up either by direct or indirect electro-oxidation process. In direct anodic oxidation method, the pollutants are made to adsorb on the anode surface and destroyed by the anodic electron transfer reaction whereas in indirect oxidation method, strong oxidants (hypochlorite/chlorine, ozone, hydrogen peroxide) are electrochemically generated in-situ and it destroys the pollutants in the bulk solution [14,15]. The generation of oxidants such as hypochlorite/chlorine in a solution having chloride ions during electrochemical process is given by the following reaction:

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (1)$$

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \quad (2)$$

$$HOCl \rightarrow H^+ + OCl^- \quad (3)$$

The practical use of mediated indirect electrochemical oxidation process by the oxidants chlorine/hypochlorite was studied for various effluents including pharmaceutical wastewater by many researchers. Dominguez et al. proved an adequate removal of total organic carbon (TOC) by employing boron doped diamond anode, which showed higher corrosion stability [16]. By performing different combination of current density and flow rate in their experiments, almost 100% TOC removal was observed. Ramesh babu et al. in their experimental study used electro-oxidation process for treating pharmaceutical effluent with NaCl as supporting electrolyte in a cylindrical flow reactor using various current densities (2–5 A/dm^2) and flow rates (10–40 L/h) [17]. It was found out that 85.56% of chemical oxygen demand (COD) reduction was obtained with flow rate of 10 L/h and applied current density of 4 A/dm^2. Ensano et al. explored the workability of treating pharmaceuticals using an intermittent electrocoagulation process and reported the significant effects of pharmaceutical degradation [18]. Maximum removals of diclofenac, carbamazepine and amoxicillin were obtained as 90%, 70% and 77%, respectively, at a current density of 0.5 mA/cm^2. Perez et al. [19] compared the treatment of real pharmaceutical effluents with Fenton oxidation (FO) and conductive-diamond electro-oxidation (CDEO) processes. They found that CDEO was found to be more effective for 80% of the samples than FO process.

In the electrochemical reactor system, the efficiency of organics degradation during electro-oxidation process depends on the residence time of wastewater in the reactor [20]. Analyzing the mixing characteristics of the fluid in the treatment system play a vital role as it affects both the efficiency of the treatment process and the hydrodynamic behavior of the reactor [21]. Studying the hydrodynamic behavior of the liquid flow helps to determine the residence time and distribution of fluid flow inside the reactor [22]. Good mixing promotes the degradation rate making the reactor system to approach ideal state [23]. In order to achieve a good electrochemical reactor design, it is important to study the flow characteristics of the fluid during the electrotreatment process. To overcome the limitation occurred in the real reactors, it is essential to design a reactor with less non-ideal effects such as fluid channeling or short-circuiting, back mixing and stagnant or dead regions. These defects decrease the efficiency of the reactor performance in either pilot plant or industrial scale [24]. All these defects in the flow dynamics can be identified by evaluating the residence time distribution (RTD) and thereby the degree of dispersion of the flow elements inside the electrochemical reactor [25].

In this work, the performance of CER for treating pharmaceutical effluent was evaluated by studying the flow dynamics of the fluid during the electro-oxidation process. The flow characterization inside the reactor was done by performing RTD studies with various flow rates such as 50, 60, 70, 80 and 90 L/h. This revealed that the flow behaviour of CER approached plug flow condition for the optimum flow rate of 70 L/h showing higher plug flow index and lower dispersion value compared with all other flow rates. The results were validated by carrying out the degradation experiments in CER using Ti/lead oxide (PbO) anode at a fixed current density of 2.5 A/dm^2 with the same flow rates (50, 60, 70, 80 and 90 L/h) and reported the reductions in COD, total dissolved solids (TDS), suspended solids (SS), pH and energy consumption during the process of electrochemical treatment.

2. Experimental

2.1. Materials

Pharmaceutical wastewater was collected from a pharmaceutical industrial unit in Chennai, India. The composition of the wastewater is determined using APHA Standard Methods [26] and are presented in Table 1.

Table 1
Characteristics of pharmaceutical process wastewater

Serial number	Factors	Raw effluent (mg/L)
1	Color	Black
2	Odor	Fouling smell
3	COD	1,120
5	TDS	24,000
6	SS	2,400
7	pH	5.15

2.2. Experimental setup

The schematic representation of a bench top CER for performing the degradation experiments is shown in Fig. 1. The experimental setup consists of two parts, one is electrolytic flow circuit consisting of reservoir, pump and flow meter, and the other one is electrical (current) flow circuit consisting of DC power supply, ammeter and voltmeter. Experiments were conducted under the fixed current density of 2.5 A/dm^2 using PbO coated on Ti mesh as anode and stainless steel acting as cathode. The diameter of the stainless steel cathode is 0.05 m and height is 0.3 m. The thickness of the cylindrical reactor is 0.002 m. The diameter of the PbO coated Ti mesh with perforation (60%) is 0.03 m and height is 0.2 m which resulted in an effective anode area of 0.01362 m^2. The anode is inserted inside the cathode and is sealed with the end frames. In the cylindrical electrolytic cell, a batch recirculation operation was performed. Electrodes are fixed in the cylindrical cell frame with a spacing of 0.02 m. The cylindrical electrochemical cell volume is measured as 0.00176 m^3. The cylindrical electrolytic cell is fixed with a rigid frame, and the electrodes are connected to a power supply made of AE Rectifier (230 V input, 0–50 V output, 100 A). DC power is supplied to the electrodes according to the fixed current density of 2.5 A/dm^2 and the experiments are carried out under constant current conditions.

2.3. Coating of anode with PbO

Thin PbO film coated on Ti electrode, commonly known as DSAs, is the most widely utilized anodes for the electrochemical treatment due to their excellent stability. These electrodes have excellent corrosion resistance and high electrocatalytic activity. The reactor setup for coating the anode is shown in Fig. 2. A cylindrical mesh made of Ti is placed inside the reactor and it is acting as anode. For coating of anode with PbO, dissolve 525 g of lead nitrate ($PbNO_3$) and 45 g of cupric nitrate ($Cu(NO_3)_2$) in distilled water and dilute to 1.5 L. The temperature is maintained at about 50°C–70°C with current density of 2.5 A/dm^2. The mixture is recirculated into the CER having Ti mesh inside using the peristaltic pump until the desired amount of coating is reached (~35 g/m^2).

2.4. Experimental procedure

2.4.1. RTD experiment

All RTD experiments were carried out in CER with water as an electrolyte and HCl acting as a tracer in room temperature condition. At different inlet flow rates (50, 60, 70, 80 and 90 L/h), water from the effluent reservoir was allowed to pass into the reactor. In the pulse input mode, 5 ml of HCl was injected into the reactor entrance in continuous operation. The time and conductivity of water were noted at regular intervals of time (30 s) at the reactor outlet. The experiment was about to end when the conductivity reduced to the level of normal water. The experimental value of exit age distribution $E(t)$ was determined to optimize the flow characterization and performance of the reactor.

2.4.2. Treatment of pharmaceutical wastewater

For treating the pharmaceutical wastewater, the experiments were performed in cylindrical electrochemical reactor (Fig. 1). Three liters of effluents per batch of electrolysis are electrolyzed by passing a fixed quantity of electricity with current density of 2.5 A/dm^2, and the process of electrolysis is carried out at various flow rates (50, 60, 70, 80 and 90 L/h) of electrolyte. To verify the destruction of COD, samples were drawn at predetermined intervals to measure the values of COD.

2.5. RTD profiles and its design parameters

The experimental determination of RTD was done by using the method of tracer response. $E(t)$ curve is obtained by dividing the concentration of the tracer $C(t)$ to its integral

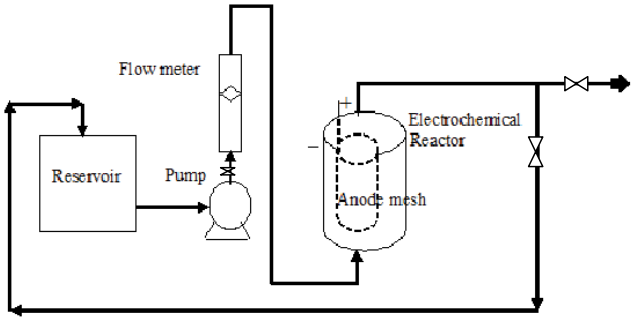

Fig. 1. Schematic representation of cylindrical electrochemical reactor setup.

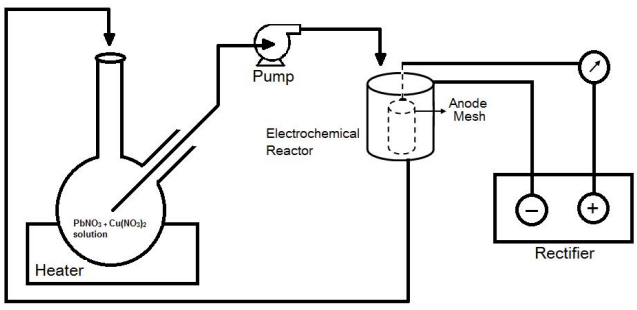

Fig. 2. Schematic representation of experimental setup for coating the electrode in CER.

at time t. Using the following equation, $E(t)$ curve can be evaluated [27]:

$$E(t) = \frac{C(t)}{\left[\int_0^\infty C(t)dt\right]} \tag{4}$$

The characteristic RTD design parameters of fluid flow characteristics inside the reactor system can be determined from the below relations:

$$t_m = \int_0^\infty tE(t)dt \tag{5}$$

$$\tau = \frac{V_r}{Q} \tag{6}$$

$$\sigma^2 = \int_0^\infty (t - t_m)^2 E(t)dt \tag{7}$$

For the closed vessel configuration, the dispersion number $N_d = \frac{D}{uL}$ can be calculated by trial and error procedure using Eq. (5), when the dispersion number is less than one [28].

$$\left(\frac{\sigma^2}{t_m^2}\right) = 2\left[\frac{D}{(uL)}\right] - 2\left[\frac{D}{(uL)}\right]^2\left(1 - e^{-\left[\frac{(uL)}{D}\right]}\right) \tag{8}$$

where σ^2 is the variance (min^2), t_m is the mean residence time (min), D is the diffusion coefficient (m^2/s), u is the fluid flow velocity (m/s), L is the length of the reactor (m), V_r is the reactor volume (L) and Q is the volumetric flow rate of the fluid (L/h).

To analyze the relationships between various RTD design parameters, such as peak time (t_p), hydraulic total residence time (τ) and mean residence time (t_m), the following relations were used [21,25]:

$$\text{Plug flow index} = \frac{t_p}{\tau} \tag{9}$$

$$\text{Dead zone index} = \frac{t_m}{\tau} \tag{10}$$

$$\text{Short circuiting index} = 1 - \left(\frac{t_p}{t_m}\right) \tag{11}$$

2.6. Analytical procedure for degradation experiments

The wastewater analyses such as pH, conductivity, COD, TDS and SS were carried out in agreement with the standard methods for examination of water and wastewater [26]. The samples withdrawn during experimental runs were titrated with concentrated sulphuric acid to arrest the variation of COD and analysis of COD is carried out as per ASTM standards. The removal efficiency of COD (R) was calculated using the following equation:

$$R = \left[\frac{Y_0 - Y}{Y_0}\right] \times 100 \tag{12}$$

where Y_0 and Y were initial and final values of COD measured during experimental runs.

The specific electrical energy consumption (E) for removing 1 kg of COD was determined using the equation below and was expressed in kW h.

$$E = \left[\frac{(IVt/\text{vol})}{\Delta COD}\right] \tag{13}$$

where ΔCOD is difference in COD between initial and final values (kg/L), vol is the reactor volume (L), I is the current passed (A), t is the retention time (h) and V cell voltage (V).

3. Results and discussion

The RTD studies were performed to study the effects of flow rates on flow characteristics of the fluid (electrolyte) inside the CER. The performance of the CER was experimentally validated by investigating the degradation of pharmaceutical effluent at various flow rates for the fixed current density of 2.5 A/dm^2. The removal efficiency of COD with energy consumption from pharmaceutical industry wastewater was studied by electrochemical method using T_i/PbO anode in the reactor.

3.1. Effect of flow rates on flow dynamics

The effect of fluid flow characteristics in CER was analyzed by evaluating various flow rates such as 50, 60, 70, 80 and 90 L/h. The effect of flow rates was found to be significant on the obtained RTDs and on the fluid flow behavior in the CER. Fig. 3 shows the exit age distribution $E(t)$ curve for various flow rates. The nonsymmetrical $E(t)$ curve shows the presence of short circuiting or bypassing along the reactor [21]. From Fig. 3, it was found that on increasing the flow rate, the peak points in $E(t)$ curve tends to increase. It reached the highest value for the flow rate of 70 L/h, which has good mixing condition and lack of back mixing characteristics. Thus, the optimum flow rate was found to be 70 L/h with indications of fluid flow behavior showing less non-ideality conditions. The $E(t)$ curve also approached near symmetrical for 70 L/h depicting much less short circuiting condition along the reactor length. Thus the flow in CER tends to approach the condition of plug flow.

Table 2 shows the various design parameters evaluated using the data of RTD experiments obtained for different flow rates in CER. It shows that upon increasing the flow rate, there is a decrease in the time spent by the fluid elements inside the reactor, and on further increase in the flow rate above 90 L/h it should have a minimal effect on the total residence time. Table 2 depicts the value of plug flow index upon increasing the flow rate of the effluent; it approached a higher value of 0.994 for the flow rate of 70 L/h. This shows that the liquid flow behavior of CER approaches to plug flow condition to a greater extent with the existence of mesh type

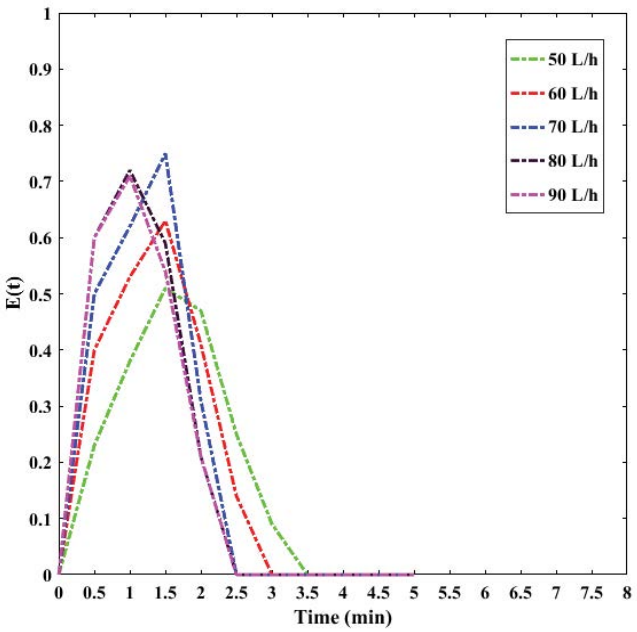

Fig. 3. Exit age distribution $E(t)$ curve for different flow rates.

cylindrical electrode. The calculation depicts the occurrence of a dead or stagnant zone at only lower flow rates (50 and 60 L/h) and the effect of stagnant zones is removed upon increasing the flow rate greater than 60 L/h. The results thus support to the evidence that packed bed reactor operating with optimum and high flow rate approach plug flow condition [24,29]. The dispersion number shown in Table 2, also, supports the results.

The fluid flow behavior in CER shows lowest deviation from plug flow [$(D/(uL) = 0.102$] only at the inlet flow rate of 70 L/h on comparing with other flow rates. The variance (σ^2) and dispersion (D) influence on the efficiency of the electrochemical reactor to degrade the effluent. RTD data discussed above have been compared with the experimental results of the degradation studies of the pharmaceutical effluent carried out with same set of inlet flow rates of the effluent. Thus the optimum values obtained to achieve plug flow behavior in CER at the flow rate of 70 L/h are shown in bold letters in Table 2. The plug flow behavior of the optimum flow rate (70 L/h) from the RTD analysis matches well with the experimental degradation studies in CER producing higher percentage of COD removal and are discussed in Sections 3.2 and 3.3.

3.2. Effect of flow rates on COD removal

Optimization of flow rate is important in investigating the efficiency of wastewater treatment in the electrochemical reactor. The effect of flow rate on the removal efficiency of COD was studied by carrying out experiments at various flow rates such as 50, 60, 70, 80 and 90 L/h at a current density of 2.5 A/dm^2. COD analysis was carried out for samples collected at regular intervals of experimental runs operated for various flow rates (50, 60, 70, 80 and 90 L/h). Keeping the initial COD of the sample in the range of 950–1,120 mg/L, electro-oxidation process was done to attain the final COD as 160–240 mg/L. Hence, the time of electrolysis was found to be varied for different flow rates to attain the final COD. Fig. 4 shows the degradation of COD for various flow rates from its initial value to a final level concerning to the electrolysis time. The COD removal efficiency was calculated for all the flow rates and are shown in Fig. 5. It was found that COD removal efficiency was found to be increasing with flow rate

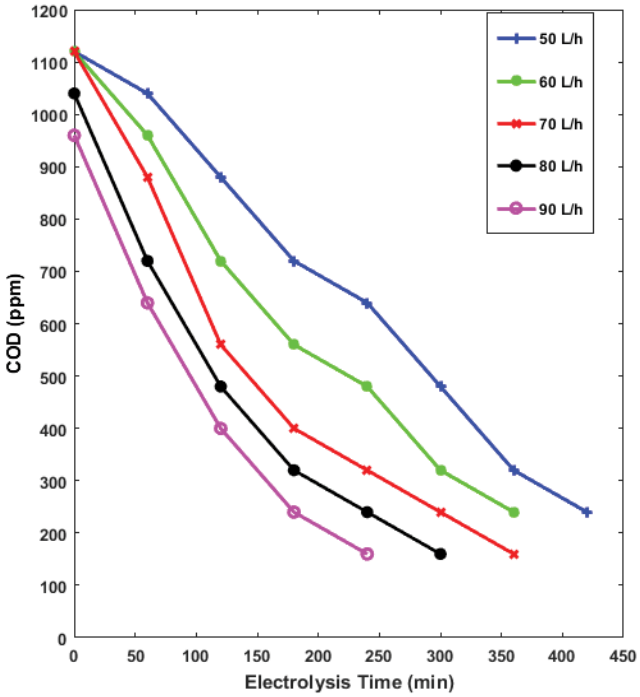

Fig. 4. COD vs electrolysis time for different flow rates.

Table 2
Parameters obtained using the data of RTD experiments

Flow rates, Q (L/h)	τ (min)	t_m (min)	t_p (min)	σ^2 (min)^2	$[D/(uL)]$	Plug flow index (t_p/τ)	Dead zone index (t_m/τ)	Short circuiting index ($1 - (t_p/t_m)$)
50	2.112	1.978	1.5	5.847	0.172	0.710	0.936	0.242
60	1.760	1.546	1.5	2.683	0.167	0.853	0.878	0.030
70	**1.509**	**1.522**	**1.5**	**1.531**	**0.102**	**0.994**	**–**	**0.014**
80	1.320	1.519	1.0	1.642	0.134	0.758	–	0.342
90	1.173	1.514	1.0	1.649	0.135	0.853	–	0.339

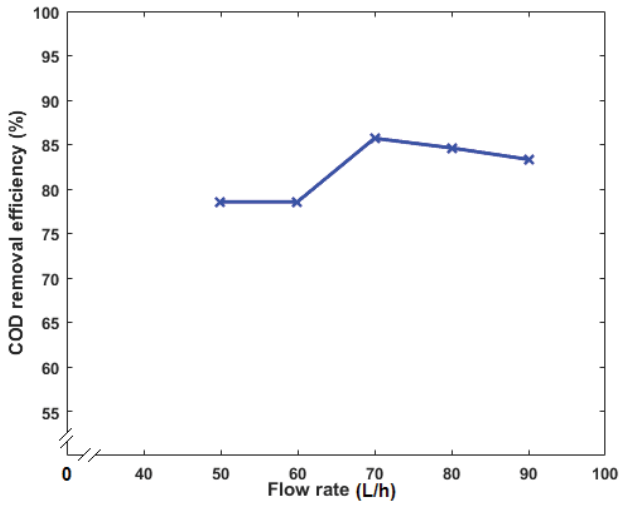

Fig. 5. COD removal efficiency vs flow rates.

until it reached the optimum flow rate of 70 L/h and then it started to decrease. The optimum removal efficiency of COD was found to be 85.71% at 70 L/h. The results showed that higher COD reduction occurred at 70 L/h at which the fluid flow characteristics of CER approaches to plug flow behavior with effective removal of short circuiting and avoiding the presence of stagnant zones to a greater extent. This has been verified by the RTD studies in CER and discussed in-depth in Section 3.1

3.3. Effect of flow rate on energy consumption

Electrochemical treatment is indubitably an intense energy approach and its efficiency is usually evaluated in terms of specific energy consumption (E). E is estimated in terms of consumption of energy utilized to degrade unit mass of organic substance (e.g., COD) in the effluent and is expressed in Eq. (13). The flow rate applied to the process directly affects the value of E. Figs. 6 and 7 enumerate the effects of flow rate on energy consumption and was found that a significant reduce in energy consumption was occurred at the optimum flow rate of 70 L/h where there is maximum COD reduction. Lowering the consumption of energy during the operation in CER reduces the operational cost of the system. E reduced to a value of 63.82 kW h/kg COD at the optimum flow rate of 70 L/h with higher COD reduction of 85.71% in 360 min of electrolysis time. This is due to the fact that the voltage consumption was less at 70 L/h at which the CER system approaches the plug flow behavior with less utilization of power.

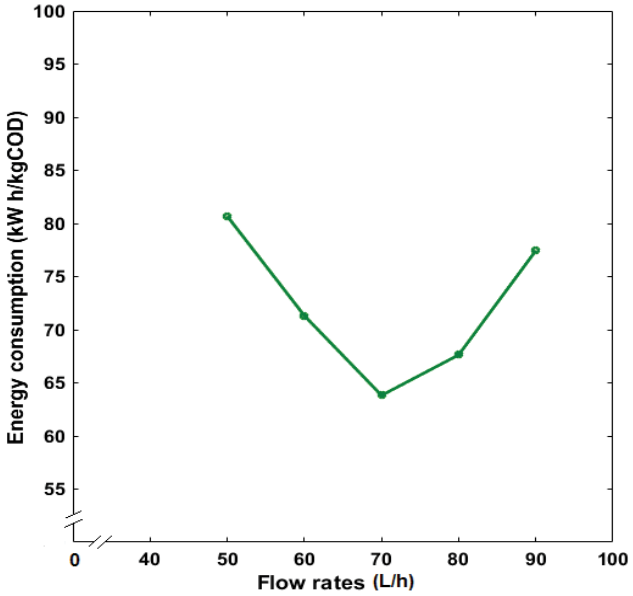

Fig. 6. Energy consumption vs flow rates.

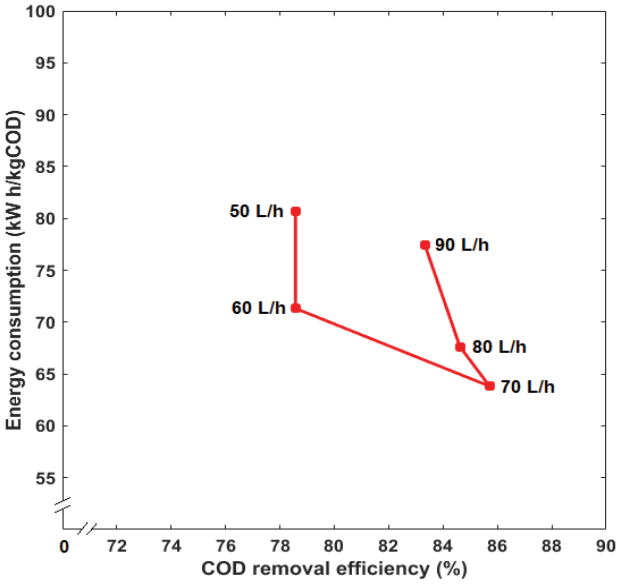

Fig. 7. Energy consumption vs COD removal efficiency for different flow rates.

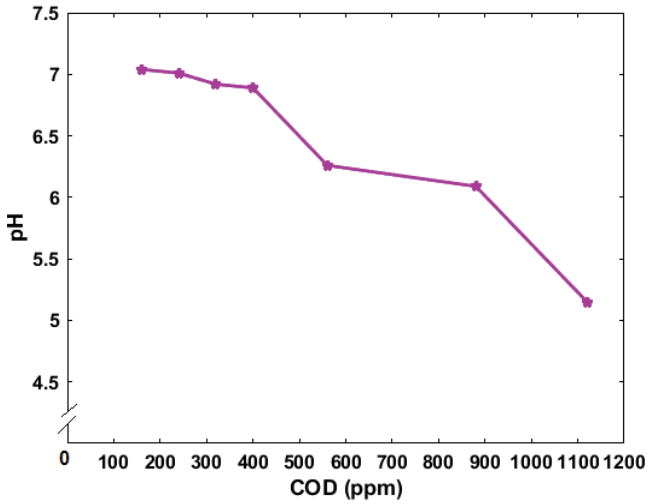

Fig. 8. Variation of pH vs COD (ppm) for the flow rate 70 L/h.

3.4. Variation of pH on COD removal

Variation in pH often controls the charge on the products of hydrolysis and metal hydroxides precipitation [30]. To analyze its variation effects during the degradation of the effluent, the pH was monitored at regular intervals and the results are shown in Fig. 8. It was found that the COD and TSS are reduced to a greater extent when the electrolysis system approached nearly neutral pH. The formation of strong hypochlorite/chlorine oxidants at neutral pH in the electrolyte removes COD at a greater extent. These oxidants are generated electrochemically in-situ and are utilized immediately for destroying the pollutants in the electrolyte by electro-oxidation process [14,15].

4. Conclusions

In this research work, the electrochemical oxidation process was investigated for the treatment of pharmaceutical effluent, and the flow dynamics were studied in the CER. The RTD method was used to understand the flow dynamic behavior of the electrolyte within the reactor for various flow rates such as 50–90 L/h. The results showed that the plug flow index approached a higher value of 0.994 and the dispersion number [$D/(uL)$] to a lower value of 0.102 for the flow rate of 70 L/h when compared with other flow rates. This shows the flow behavior of CER approaches to plug flow condition to a greater extent for the optimum flow rate of 70 L/h. The calculation also depicts the presence of a dead zone only at low flow rates (50 and 60 L/h) and on increasing the flow rate to more than 60 L/h removes the effect of stagnate and dead zones. The RTD observations have been compared with the experimental results of the degradation studies of the pharmaceutical effluent carried out with same set of inlet flow rates of the effluent. The plug flow behavior of the optimum flow rate (70 L/h) from the RTD analysis matches well with the experimental degradation studies in CER producing higher percentage of COD removal (85.71%) under the current density of 2.5 A/dm^2. The power consumption was found to be minimum at this flow rate with the effective removal of color, odor, TDS (91.7% removal) and SS (95% removal) with in the short residence period of effluent in the reactor. This technique can be utilized for treating different organic pollutants with high COD values that are expelled to the environment.

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