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Pulp and Papermill Effluent Treatment by Continuous Electrocoagulation

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Abstract

The continuous electrocoagulation (CEC) technique, which uses aluminium (Al) as an electrode material, is examined in this work for the treatment of highly toxic pulp and papermill effluents. The process optimization was done with respect to the flow rates (L h^{-1}) and residence time (τ) of wastewater which resulted in the excellent removal of Total solids (TS), colour and Chemical Oxygen Demand (COD). The removal rate of COD was 94.4 and 93.1% at the optimal flow rates of 0.5 and 1.0 dm^3 h^{-1}, correspondingly, whereas the colour removal rate was 91.6 and 83.6%. There was gradual decrease in the pH of the effluent during the process due to the production of aluminium hydroxides in variable oxidation states. Variation in the cell voltage during the electrolysis has also been studied. TDS of effluent was also decreased by a factor of 93% after 4 h residence time at a flow rate of 0.5 dm^3 h^{-1}. With a variation in flow rates, electrode degradation and consumption were also examined.

Keywords

Papermill, colour, Electrocoagulation, Effluent treatment, Flow rate

1. Introduction

The pulp and paper (P&P) industries consume large amounts of water and natural resources, and they release a wide range of liquid, gaseous, and solid wastes into the environment that are extremely hazardous to both human health and the environment. The effluent has a dark colour, a bad odour, a high organic content, and extremely high levels of COD, BOD, and pH.

High organic matter and suspended material concentrations are often regarded as important contaminants of these effluents. Chemical pulping, in particular, produces high-strength effluent that contains suspended solids, organic material, and wood debris, as well as soluble wood components. Furthermore, the usage of chlorine for bleaching pulp creates a large number of harmful compounds since chlorine is needed to brighten the pulp. The wastewater produced by the pulping process contains a variety of wood chemicals such as lignin, carbohydrate, and extractives. Chlorination is often the initial stage in kraft bleaching, and chlorinated organic compounds are created during this process [1]-[4].

To reduce pollutant load and adhere to environmental standards, pulp and paper mill wastewater must be treated. For the treatment of pulp and paper industry wastewater, several approaches have been used, including physical, chemical, electrochemical, and biological processes [5]-[7]. Electrocoagulation is a promising approach for treating wastewater. It incorporates the benefits of electrochemistry, floatation, and agglomeration. Electric current is used in an electrochemical procedure to eliminate dissolved, emulsified, or suspended contaminants from water. Because it is an environmentally benign choice that can create the least sludge, requires no chemical additions, and has the smallest impact without compromising the water quality after treatment, electrochemical wastewater treatment technology has started to receive prominence. The fundamental distinction between chemical coagulation cum flocculation and electrocoagulation is the usage of chemical coagulants and flocculants. A suitable anode material is electrolytically oxidized to produce the coagulants in situ throughout electrocoagulation, which produces substantially less sludge [8]-[9].

Type of electrodes, gap between electrodes, number of electrodes, size of electrodes, configuration of metals, current density, charge loading, pH of sample, addition of supporting electrolyte, and duration of operation are a few factors that affect the effectiveness of electrocoagulation [10]. Iron, aluminium, and stainless steel are the most frequently used electrodes. These metallic plates serve as anodes and cathode in the electrolytic cell of electrocoagulation process. These pairs of metal sheets are known as anode and cathode. During the process, sacrificial anode is oxidized (loses electrons) and the water is reduced (gains electrons) at cathode resulted in treated water and easy to settle floc [11].

The removal of COD, colour, suspended particles along with change in pH is altered by the use of several electrodes. Coagulants are generated from electrodes (aluminium). As per the reactions, dissolution of anode (aluminium) produces cations. As the counter ions create coagulation, positive-charged metal ions are drawn to the negatively charged hydroxides in this solution, producing ionic hydroxides that have a higher attraction to the dispersed particles [8].

$$Al(s) \rightarrow Al^{3+}(aq) + 3e^{-} \quad \text{(i)}$$

$$2H_2O + 2e^{-} \rightarrow H_2 + 2OH^{-} \quad \text{(ii)}$$

$$Al^{3+}(aq) + 3OH^{-} \rightarrow Al(OH)_3 \quad \text{(iii)}$$

The arrangement of electrodes in the reactor is a critical factor that can impact the performance of the electrocoagulation process. The electrodes can be arranged in either a series or parallel configuration. In the monopolar parallel setup, all anodes are connected to each other, and similarly, all cathodes are also connected. In the monopolar series arrangement, the outermost electrodes are connected to a power source, and the current then passes through the remaining electrodes, polarizing them. The electrodes connected in series mode experience higher resistance, thus requiring higher potential for a given current. Maintaining the framework is simpler in the bipolar parallel connection due to its easier setup [9].

Previous studies have reported the successful use of Electrocoagulation for distillery wastewater treatment, but no data has been found regarding its use for pulp and papermill wastewater. This study is conducted using a parallel electrodes framework for electrolysis. The current study focusses on the investigation of the efficacy of the continuous electrocoagulation (CEC) process for treating pulp and papermill wastewater. It focuses on testing the efficiency of the process under various working parameters such as pH, cell voltage, flow rate, and residence time. Furthermore, it explores the impacts of the residence time and flow rates for the reduction of COD, Total Solids, turbidity and colour. The dissolution and disintegration

rate of the anodes (aluminium) have also been explored with the CEC process. Section 2 comprises the experimental setup of CEC process, chemistry of electrocoagulation, analytical methods used for the determination of various physicochemical attributes of effluent before and after treatment. Section 3 describes in detail the effect of electrocoagulation procedure on the COD, colour, pH, Total dissolved solids and anode disintegration, etc.

2. Materials and Methods

The effluent samples of wastewater were collected from Suchi paper mill, Ghaziabad (28.6169° N, 77.4777° E), India. Collection of effluent was done in PET bottles which were then kept at 4°C in refrigerator till used. The wastewater was translucent, dark brown in color (due to lignin and tannin derivatives). Analytical reagent grade chemicals were used in the present research work.

2.1. Experimental arrangement

Figure 1 illustrates the experimental setup for the CEC treatment of pulp and papermill effluent. Various CEC trials were conducted on the effluent under different operational conditions such as flow rates and residence times. To maintain a consistent concentration of the feed, the feed chamber and CEC apparatus were continuously stirred by magnetic stirrers. A peristaltic siphon was utilized to establish the feed flow rate to the CEC apparatus. The cell voltage was measured using a digital multimeter. Treated samples were obtained from the outlet of the CEC apparatus at regular intervals, filtered, and analyzed for color and COD reduction.

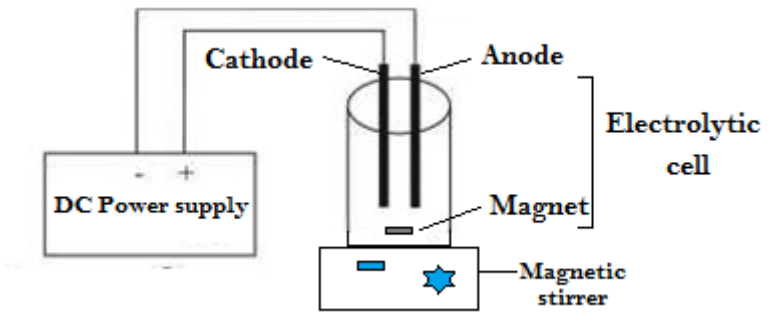

Figure 1. Schematic diagram for the Experimental arrangement of Electrocoagulation Process

2.2. Analytical Methods

Digital pH meter consisting of combined glass electrode was used to measure pH of samples. Reflux and titrimetric methods were used to determine the COD of samples. The color intensity was evaluated by using a spectrophotometer (Perkin Elmer, Switzerland) as per standard methods given by (APHA) [12].

2.3. Chemistry of Electrocoagulation

The production of Al^{3+} and hydroxide ions occurs through electrochemical degradation of the aluminium electrodes. This redox process is intricate and involves several steps, including hydrolysis, polymerization, and precipitation [13]. Through these processes, three types of products are formed (i) low molecular weight aluminium hydroxides (b) hydrolytic aluminium polymer and (c) precipitated aluminium oxides. The further hydration of Al^{3+}ions depend on the pH of the solution.

3. Results and Discussion

Physicochemical parameters *such* pH, TS, TSS, TDS, BOD, COD, etc. of pulp and paper mill effluent were measured and the readings are reported in Table 1.

Table 1: Physicochemical analysis of pulp and paper mill effluent

Parameters	Mean value
Odour	Unpleasant
pH	7.9
Total suspended solids (TSS)	9,566.66 (±88.19)
Total dissolved solids (TDS)	17686.66 (±566.10)
Total organic carbon	2880(±22.30)
Colour	6280.34 (± 97.85)
BOD	32000± (577.35)
COD	45000 (±946.48)

3.1. Variation in COD upon Electrocoagulation

The maximum drop in the COD values of the effluent was observed at the slowest operational flow rate and highest residence time (τ). The COD removal rate of 94.6% was measured when τ was 4 h and flow rate was 0.5 $dm^3\ h^{-1}$ whereas when τ was 1 h and flow rate was 4 $dm^3\ h^{-1}$, the removal rate was very poor of about 10% (Fig. 2). It has been seen that as the residence time in the Combined electrocoagulation device reduces, there is slow formation of a thick precipitate. At τ < 1 h, white pearl-coloured scales of aluminium oxides begin to deposit on anodes which slashes down the current by 0.06 A. At lower τ (⩽ 0.6 h), the suspended precipitate particles were found to stick on the edges of anode. As the process proceeds further, precipitate particles begin to behave as the nucleus for additional agglomeration, covering the entire plate region by curdy precipitates [9].

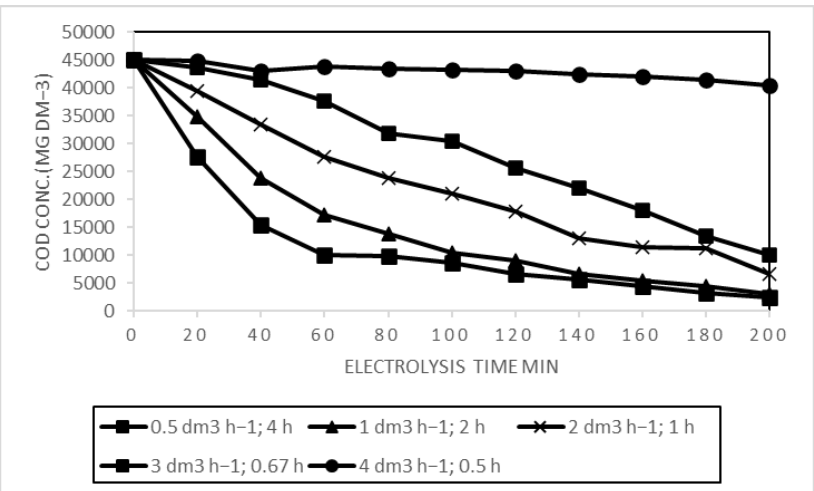

Figure 2. Variation in COD of pulp and papermill effluent at different flow rates and residence times

3.2. Variation in Colour intensity upon Electrocoagulation

Reduction in color intensity of the pulp and papermill effluent wastewater at various residence time is shown in Fig. 3. There was an unparalleled increase in color intensity from about 6000 to about 10000 during the early 20 minutes of the process. This finding can be clarified by considering the fact that in the beginning of the process there was oxidative polymerization of pulp and papermill effluent which leads to the formation of organic compounds that are dark brown inAfter this point, the color drop begins, and which is directly related to the electrolysis time. The maximum colour removal rate was 91.6% at τ = 4 h whereas at τ = 0.5 h it was just a scanty of 13.6%.

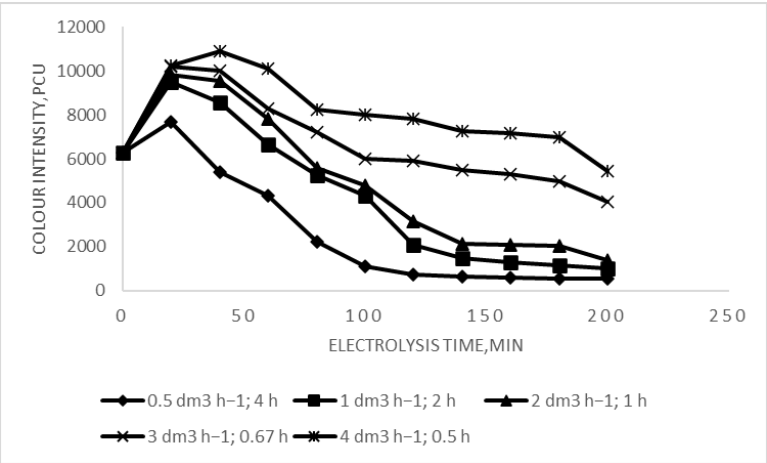

Figure 3. Variation in color intensity of pulp and papermill effluent at different flow rates and residence times

3.3. Variation in pH upon Electrocoagulation

Variation in the pH of the pulp and papermill effluent with electrolysis time at various flow rates and residence times is shown in Fig 4. There was a gradual decrease in the pH of the effluent during the course of the process. The pH of the treated effluent ranges from 6.59 to 7.40 at different flow rates and residence times. There was a direct relationship of decline in pH of effluent with the electrolysis period. The phenomenon could be explained by the emergence of aluminium hydroxides with different charges such as $Al(OH)^{2+}$, $Al(OH)_2^{+}$ and Al(OH)3. The maximum decrease in the pH was observed with the electrolysis time of 4 h at the flow rate of 0.5 $dm^3\ h^{-1}$.

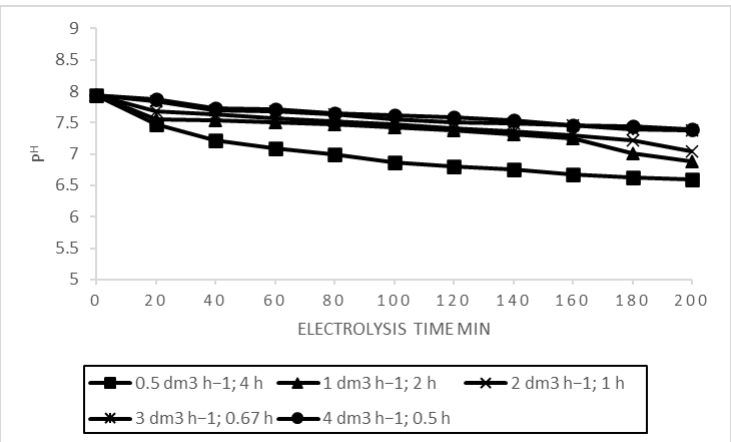

Figure 4. Variation in pH of pulp and papermill effluent at different flow rates and residence time

3.4. Variation in Cell voltage upon Electrocoagulation

Variation in cell voltage throughout electrolysis process for different flow rates is presented in Fig. 5. Regardless of the flow rates, the initial cell voltage, which was between 3.3 and 3.4 V, started to decline as the process went on. After 60 minutes of electrolysis at the flow rates of 2 L h 1, the cell voltage begins to climb once more gradually and eventually becomes very near to the starting value. At this time, tiny flakes start to develop at anode, which is in conjunction with a decrease in colour, COD, and other complex organic compounds. It is worth noticing that the cell voltage keeps decreasing and doesn't reach its initial value at flow rates greater than 3 L h^{-1}. Large scales of precipitates are formed during this period, which covers the entire surface of anode causing reduced removal effectiveness [12].

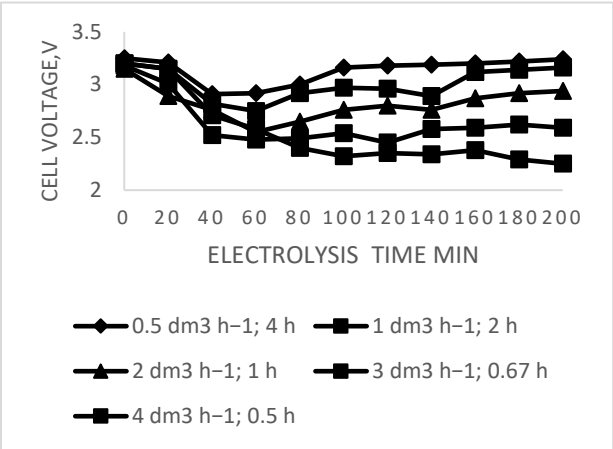

Figure 5. Change in Cell voltage of pulp and papermill effluent at different flow rates and residence times

3.5. Variation in Anode Consumption and Total Solids concentration upon Electrocoagulation

The variation in the amount of anode consumption (g) and concentration of total solids (TS; g/L) is shown in Fig. 6 at flow rate of 0.5 L h^{-1} and residence time of 4 h for different electrolysis time. With the increase in τ, both the quantity of anode dissolution and the volume of sludge generation increased. Initially, there was an increase in total dissolved solids (TDS) concentration which could be credited to oxidative dissolution of anodic aluminium into the pulp and papermill effluent. However, as the procedure went on, precipitation of aluminium hydroxides and other organic species started, which decreased the TDS. At the residence time of 4 h and flow rate of 0.5 L h^{-1}, the decrease in TDS of effluent was found to be ~16.1 g/L (93%).

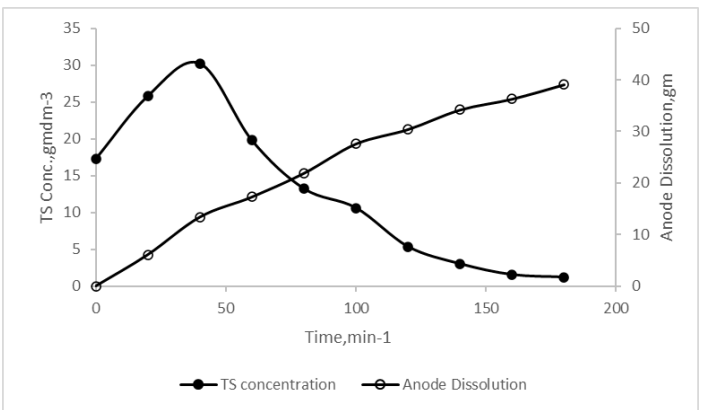

Figure 6. TS concentration and anode disintegration versus Electrolysis time at the flow rate of 0.5 Lh^{-1}

4. Conclusions

When compared to conventional techniques of treatment, the current investigation of continuous electrocoagulation of pulp and papermill effluent shows that process generates a significant decrease in the pollution load of the effluent. It has been investigated how varied flow rates and eletrolysis times affect the colour, COD, pH, and TDS of pulp and papermill effluent. According to the research, the ideal flow rate and residence time for this electrocoagulation process are 0.5 L/h and 4 h, respectively. According to the experimental findings, the effluent's colour (91%), COD (94%) and total dissolved solids (93%) have all significantly decreased. In pulp and papermill effluent, the initial complex organic molecules such lignocellulosic derivatives, chlorinated phenolic compounds, and adsorbable organic halides are converted into simple inorganic compounds after electrocoagulation treatment, which lowers the colour, COD, and total dissolved solids. The pH of the effluent decreased from the basic to the slightly acidic side, making it appropriate for discharge in water bodies. The removal effectiveness demonstrates that electrocoagulation is a safe method for treating pulp and papermill wastewater.

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