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Studies on various operating parameters for the removal of COD from pulp and paper industry using electrocoagulation process

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ABSTRACT

This study describes a batch electrocoagulation process for treating pulp and paper industry wastewater using iron plates. The effect of various operating parameters such current density, initial effluent pH, initial COD concentration, electrode combination and interelectrode distance on the removal of percentage COD and power consumption were investigated. The experimental results showed that 85% COD removal with 5.16 kWh/m^3 of power consumption was observed for initial COD concentration of 2,500 ppm with current density of 0.40 A/dm^2, initial effluent pH of 7.0 with interelectrode distance 1 cm at the end of the 120 min of operation. It was observed that, an increase in current density, decrease in the effluent COD concentration, Fe/Fe electrode combination, and decrease in interelectrode distance improved the COD removal efficiency. Optimum initial effluent pH for highest COD removal was 7.0. Furthermore in this investigation, extraction of lignin from the pulp and paper industrial effluent also carried out and analyzed by FT-IR system.

Keywords: Pulp and paper industry; Lignin; Electrocoagulation; COD and power consumption

1. Introduction

The rapid increase in population and the increased demand for industrial establishments to meet human requirements have created problems such as overexploitation of available resources, leading to pollution of the land, air, and water environments. The pulp and paper making industry is a water-intensive industry; it is ranked third based on water consumption after metals and chemical industries. The Indian pulp and paper industry consuming 100–250 m^3 freshwater/ton of paper production depends on the process [1]. The wood pulping and production of the paper products generate a considerable amount of pollutants characterized by biochemical oxygen demand (BOD), chemical oxygen demand (COD), suspended solids, toxicity, and color when untreated or poorly treated effluents are discharged to receiving waters [2].

The significant environmental impacts of the manufacture of pulp and paper industries result from the pulping and bleaching processes. In some processes, sulfur compounds and nitrogen oxides are emitted to the air, chlorinated and organic compounds, nutrients, and metals are discharged to the wastewaters [3]. The high water usage, between 20,000 and 60,000 gallons per ton of product, results in large amounts of wastewater generation. The effluents from the industry cause slime growth, thermal impacts, scum formation, color problems, and loss of esthetic beauty in the environment [4]. The characteristics of the wastewater generated from the pulp and paper industry depend upon the types of raw material, types of process technology applied, and internal recirculation of the effluent and management practices. In order to select and design a suitable treatment technique for reuse of effluent in the process application, a complete knowledge of characteristics of the particular industrial effluent is necessary. Effluent from pulp bleaching units (80–100 m^3/ton paper production) contains dissolved lignin, carbohydrate, color, COD, AOX, and inorganic chloride compounds. The wastewater from paper making section contains particulates, organic compounds, inorganic dyes, etc.

Physical, chemical, and biological processes are quite expensive and generate a considerable amount of sludge which itself needs further treatment [5]. The low biodegradability index of pulp and paper effluent from bleaching process clearly shows that the biochemical method cannot be used [6,7]. Several methods have been considered for the removal of pollutant from the pulp and paper industrial effluents. The main treatment processes are primary clarification by sedimentation or flotation and secondary treatment by activated sludge or anaerobic digestion.

In recent years, electrochemical technologies such as electrocoagulation (EC), electroflotation, and electrooxidation have been used for the treatment of pulp and paper effluent. Among these methods, EC emerges as one of the promising techniques due to its unique feature such as complete degradation of pollutants, less sludge generation, and ease in operation. EC is a complex and interdependent process where the generation of coagulants takes place in situ by dissolving sacrificial anode.

EC is an electrolytic process consisting in the dissolution of sacrificial anodes made out of iron or aluminum upon application of a current between two electrodes for treatment of liquid wastewater containing inorganic or organic matter. As a matter of fact, Al^{3+} or Fe^{2+}/Fe^{3+} ions generated by the dissolution form neutral hydroxide complexes which act as coagulating agents for the suspended pollutants. With an iron anode, $Fe(OH)_n$ with $n = 2$ or 3 is formed. Two mechanisms have been suggested for this production [8]: Either $Fe(OH)_2$ is formed at the anode, while hydrogen is produced at the cathode from water, or $Fe(OH)_3$ is formed at the anode while hydrogen is produced at the cathode from H^+ ions. It is often considered that the cationic charges produced by the dissolution neutralize the suspended impurities which are negatively charged for $pH > 3$: The neutral entities formed are destabilized and can coalesce to larger and separable agglomerates [9].

EC is known for its effectiveness in treating charged effluents. In recent years, EC has been successfully tested to treat various charged wastewaters. It can be used for many purposes such as removal of heavy metals [10], treatment of textile effluents [9,11] or aircraft industry effluent [12] and has been widely tested to treat pulp and paper mill effluents [13–18].

From the literature review, it has been found that most of the researchers successfully used EC method for the treatment of black liquor from pulp and paper industries [19]. However, very limited works have been reported for the treatment of pulp bleaching effluent using EC method, especially using Fe electrodes and only few works had been studied for the pollutant removal using real effluent. The most of the literature work has focus on treatment efficiency based on the percentage color and/or COD removal (Table 1). In all electrochemical processes, the power consumption having important parameter due to operating costs and pollutant removal, and hence in the present investigation, we have attempted to study the influence of operating parameters such as current density, initial effluent pH, initial COD concentration, electrode combination, and interelectrode distance on the percentage COD removal and power consumption for the pulp and paper industrial effluent using EC process.

2. Materials and methods

2.1. Materials

The effluent was collected from a pulp and paper industry nearby Erode district, Tamil Nadu, India. The physicochemical characteristics of the effluent were determined before treatment. The main characteristics of the effluent were given in Table 2. All the chemicals used were supplied by Merck Specialties Private Limited.

2.2. Methods

The experimental set-up of the electrochemical reactor operated for EC consisted of a cylinder made of Perspex glass with a net volume of 600 ml (Fig. 1). An undivided electrochemical cell with a pair of electrode and DC rectifier constituted the electrochemical oxidation unit. The iron electrodes were used as anode and cathode, respectively. The anode and cathode were placed vertically and parallel to each other with an interelectrode gap of 1 cm. The volume of effluent taken was 500 ml in the electrochemical reactor, and the electrodes were connected to a regulated electrical

Table 1
EC process reported in the literature for treatment of pulp and paper industry wastewater

Types of electrode	Removal efficiency (%)	Operating time	Optimum conditions	Literature
Al and Fe	Dissolved organic carbon removal ranged between 24 and 46%, and COD removal ranged between 32 and 68%	90 min	Current density (100 and 150 A/m^2)	[20]
Al	Lignin—92%, Phenol—93%, BOD-80%, COD—55%	Electrolysis time of 2 min	Current intensity of 77.13 Ma, at 12 V	[21]
Fe	97%	Treatment time of 60 s	Initial pH—5, initial concentration of 125 mg/l, 2 A current density	[22]
Al	Color—94%, COD—90%, and BOD—87%	Electrolysis time—30 min	15 mA/cm^2 current density, pH of 7, 1 g/l NaCl, 100 rpm, 28℃ temperature and 3 cm electrode distance	[23]
Fe	Black liquor color—100%, COD—91%	–	Current density of 55.56 A/m^2 at neutral pH with a six-plate arrangement	[24]
Al and Fe	Color—99%, COD—98%	T = 50 min	(Initial pH 7, t = 50 min and J = 14 mA cm^{-2})	[25]

Table 2
Physicochemical characteristics of pulp and paper wastewater

Characteristics	Range
pH	13–14
COD	50,000 mg/L
BOD	3,800 mg/L
TDS	11,533 mg/L
Color	Dark brown
Odor	Burnt sugar

DC power supply (APLAB Ltd; Model L1606). The desired current density was applied to anode and cathode submerged in the effluent. The cell voltage and current were read periodically using a multimeter. The solution was continuously stirred at a constant speed using a magnetic stirrer. The pH of the solution was measured using pH meter (Elico; Model LI120). During the studies, the initial pH was adjusted using a reagent of 0.1 N NaOH and 0.1 N H_2SO_4. The samples were collected periodically from the reactor and filtered using Whatmann 42 filter paper. The COD was determined to investigate the behavior of EC process of pulp and paper industry wastewater.

The electrode plates were cleaned manually by washing in distilled water prior to every run, and owing to their sacrificial nature, they were replaced after every 2 runs. The COD removal efficiency and power consumption of the electrochemical reactor were studied under various conditions such as current density, initial pH, initial COD concentration, electrode combination, and interelectrode distance.

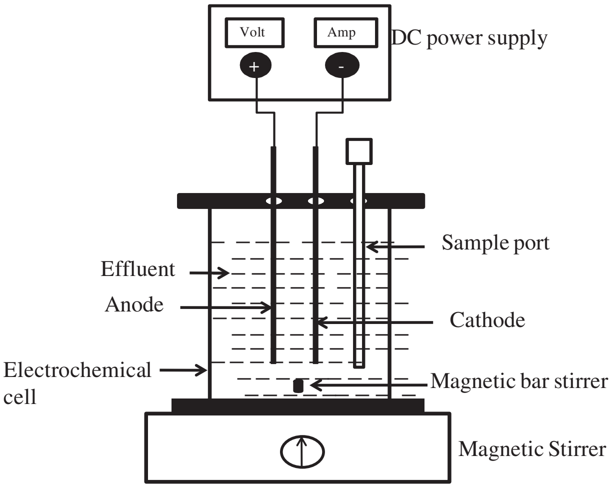

Fig. 1. EC process for the treatment of pulp and paper industry.

2.3. Analysis

2.3.1. Extraction of lignin from the pulp and paper industrial effluent

Twenty milliliter of the raw effluent was taken in a centrifuge tube, and pH was adjusted accordingly from 3 to 11 using 1 N H_2SO_4 and NaOH. The tubes were left to stand for about 20 min, thereby allowing the lignin to precipitate out. After 20 min, the samples were centrifuged at 8,000 rpm for 15 min. The supernatant was discarded in each case, and the cake was washed with acid–water. All samples were then completely dried in an oven at −60℃ prior to measuring the weight and then analyzed using FT-IR [26].

2.3.2. Determination of COD

The concentration of pollutants in the sample was measured in terms of COD. The COD value was determined by the dichromate closed reflux method strictly following the APHA.

2.3.3. Principle of COD

The organic matter present in the sample gets oxidized completely by $K_2Cr_2O_7$ in the presence of H_2SO_4 to produce CO_2 and H_2O. The excess $K_2Cr_2O_7$ remaining after the reaction is titrated with ferrous ammonium sulfate. The dichromate consumed gives the O_2 required to oxidation of the organic matter.

2.3.4. Percentage color removal

The percentage color removal was calculated by Eq. (1):

$$\text{Percentage color removal} = \frac{\text{Abs}_i - \text{Abs}_t}{\text{Abs}_i} \times 100 \qquad (1)$$

where

Abs_i and Abs_t are the absorbance of initial and at any time t samples for corresponding wavelength $\lambda_{\max}$.

2.3.5. Percentage COD removal

The percentage removal of COD in the effluent was calculated using Eq. (2):

$$\text{Percentage COD removal} = \frac{\text{COD}_0 - \text{COD}_t}{\text{COD}_0} \times 100 \qquad (2)$$

where

COD_0 and COD_t (in ppm) are the COD at time $t = 0$ (initial) and at t (reaction time), respectively.

2.4. Power consumption

Effluent treatment techniques must be technically and economically feasible. Electrical energy consumption or power consumption is a very important economical parameter in electrochemical processes. The operating cost is mainly associated with electrical energy consumption.

Electrical energy consumption for the EC process can be calculated using the equation:

$$\text{EEC}_{\text{EC}} = \frac{VIt}{V_{\text{R}}} \qquad (3)$$

V represents the observed cell voltage (V), I is the current (A), t is electrolysis time (h), and V_{R} is the volume of the effluent (L).

3. Results and discussion

3.1. Various operating parameters

3.1.1. Effect of current density

The effect of current density on the removal of percentage COD and power consumption in the pulp and paper industrial effluent with an initial COD concentration of 2,500 mg/L of the solution was studied at initial solution pH 7, and the results were shown in Fig. 2. From Fig. 2 shows that the removal of percentage COD and power consumption increased from 40 to 93% and from 1.434 to 5.894 kWh/m^3 with an increasing current density from 0.10 to 0.50 A/dm^2. This can be attributed due to fact that the applied current density determines the rates of coagulant and bubble production, which in turn can influence the process efficiency. An increase in the gas bubbles density with reduction in their size enhances upwards flux resulting increased pollutant degradation and sludge floatation [27]. However, it is advisable to limit

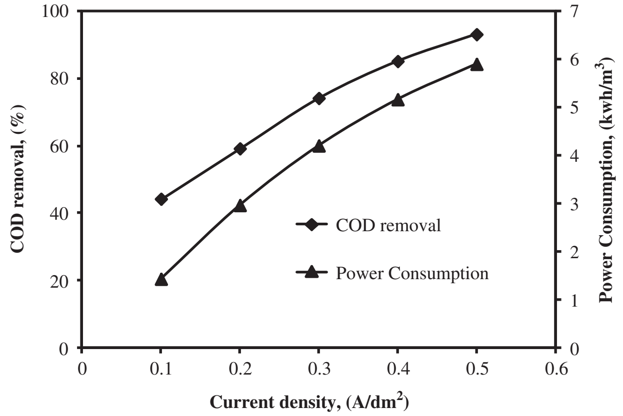

Fig. 2. Effect of current density on percentage COD removal and power consumption (effluent COD concentration: 2,500 mg/L, initial effluent pH: 7, time: 2 h, electrode combination: Fe/Fe, inter distance between the electrodes: 1 cm and stirring speed: 100 rpm).

the current density in order to avoid excess oxygen evolution as well as to eliminate other adverse effect, like heat generation and also unwanted reaction occurs. According to operating cost and removal efficiency of pollutant, we should operate at optimum current density level [28].

3.1.2. Effect of initial effluent pH

The effect of effluent initial pH plays an important role on the performance of the electrochemical process. The effect of initial effluent pH on the removal of percentage COD and power consumption for the pulp and paper industry effluent by EC process was shown in Fig. 3(A). It can be noticed from the figure, the removal of percentage COD and power consumption increased from 25 to 85% and from 1.878 to 5.16 kWh/m^3 with increasing initial effluent pH from 3 to 7. With further increasing of initial effluent pH from 7 to 11, the removal of percentage COD and power consumption decreased from 85 to 58.65% and from 5.16 to 5.37 kWh/m^3. This may be due to the decrease in the conductivity of the electrolyte when pH changes from acidic to neutral and its increase when pH changes from neutral to alkali condition in the EC process. The maximum removal of percentage COD was observed at pH 7. When pH is increased above 7, the amount of hydroxide ions was increased in solution. Consequently, some of the hydroxide ions may be oxidized at the anode. This reduces the production of iron ions, thereby decreasing the removal of percentage COD in the effluent. At lower pH, the protons in the solution were reduced to H_2 at the cathode and the same proportion of hydroxide ions cannot be produced. At lower pH, $Fe(OH)^{2+}$ and $Fe(OH)_2^+$ were produced which are disadvantageous for colorant precipitation [29,30]. In summary, higher COD removal efficiency was obtained in neutral media, as reported by several authors [31].

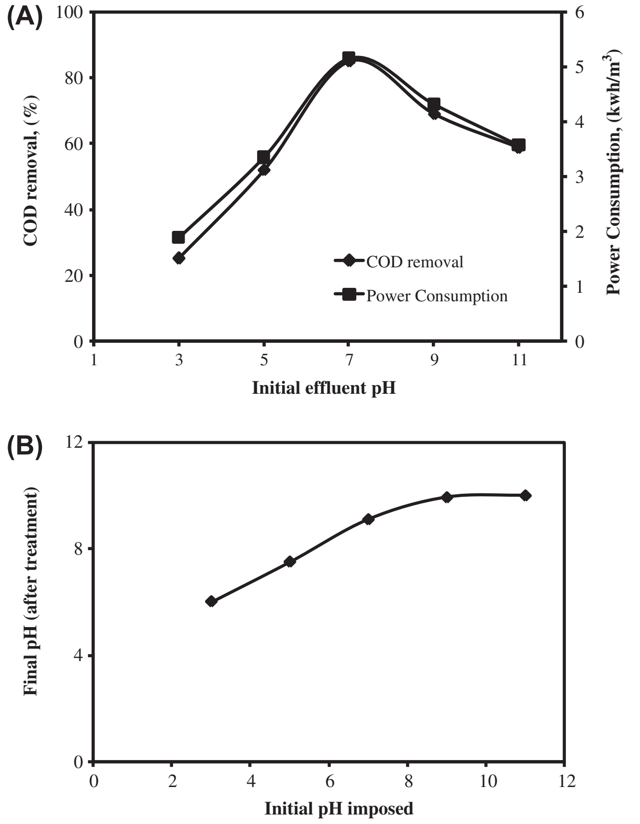

Fig. 3. Effect of initial effluent pH on (A) percentage COD removal and power consumption and (B) final pH (effluent COD concentration: 2,500 mg/L, current density —0.40 A/dm^2, time: 2 h, electrode combination: Fe/Fe, interdistance between the electrodes: 1 cm and stirring speed: 100 rpm).

Fig. 3(B) shows that the pH change of pulp and paper industry effluent after EC process. For initial pH values imposed from 3.0 to 5.0, the final values increased and stabilized around 6.0 and 7.5. However, the pH values recorded after EC and increased rapidly from 9.1 to 10.0 while the initial effluent pH value of pulp and paper industry ranged from 7.0 to 11.0. To increase in pH during EC process was primarily attributed to the evolution of hydrogen gas at the cathode and the concentration hydroxyl ions (OH^-) increases in the solution due to the electrochemical reactions.

$$2H_2O_{(l)} + 2e^- \leftrightarrow H_{2(g)} + 2OH^-$$

3.1.3. Effect of initial COD concentration

To determine the effect of initial COD concentration on the removal of percentage COD and power consumption were shown in Fig. 4. From the figure the initial COD concentration increasing from 1,250 to 5,000 mg/L decreased the removal of percentage COD from 98 to 52% and power consumption from 6.8 to 3.9 kWh/m^3, respectively. One of the most important pathways of pollutant removal by EC is adsorption of organic molecules on metallic hydroxide flocs. According to Faraday’s law, a constant amount Fe^{2+} is passed to the solution with same galvanostatic current density and electrolysis time for all initial COD concentration. Thus, same amount of ferrous hydroxyl ions could be produced in the solution. As per the results, the adsorption capacity of ferrous hydroxyl ions is limited and specific amount of ferrous hydroxyl ions is able to adsorp specific amount of organic molecules. So, the ferrous hydroxyl ions produced at high COD

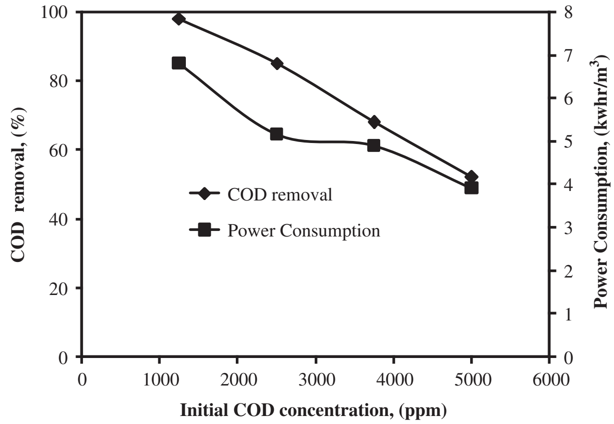

Fig. 4. Effect of COD concentration on percentage COD removal and power consumption (current density: 0.40 A/dm^2, Initial effluent pH: 7, time: 2 h, electrode combination: Fe/Fe, interdistance between the electrodes: 1 cm and stirring speed: 100 rpm).

concentration were insufficient to absorb all of the COD concentration in the solution which decreases the removal of percentage COD [32].

3.1.4. Effect of electrode combination

In EC process, mostly widely used as sacrificial anodes such as iron and aluminum, the electrodes are cheap, readily available, and proven effective. The different electrode material and combination are very important and also heart of the electrochemical process [31,33,34].

Iron and aluminum plates in four different combinations were investigated in this study to determine the better electrode combination. Fig. 5(A) and (B) illustrate the effect of different electrode combination on the removal of percentage COD and power consumption for the pulp and paper industry. As can be seen in this figure, the COD removal efficiency and power consumption were achieved in approximately 85, 62, 70, and 57% and 5.16, 5.8, 7.16, and 6.334 kWh/m^3 for Fe/Fe, Al/Fe Fe/Al and Al/Al electrode combinations, respectively. It also indicated that the Fe/Fe electrode combination yielded the best COD removal efficiency with lower power consumption, followed by the Al/Fe, Fe/Al, and Al/Al pairs. Fe and Al electrode can be explained by the chemical reactions taking place at the aluminum and iron electrodes and the reaction as follows:

For Al electrode

$$\mathrm{Al} \rightarrow \mathrm{Al}^{3+} + 3e^{-}$$

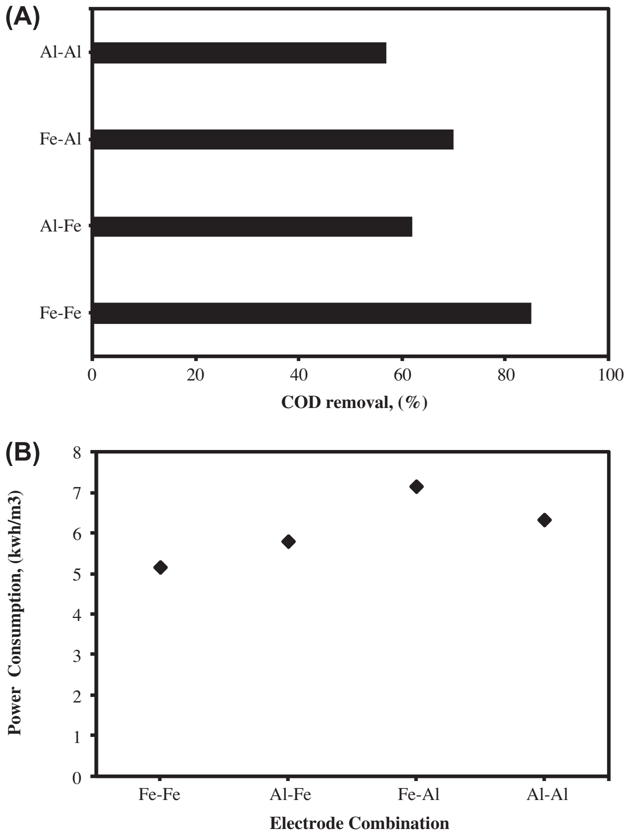

Fig. 5. Effect of electrode combination on (A) percentage COD removal and (B) power consumption (effluent COD concentration: 2,500 mg/L, current density: 0.40 A/dm^2, initial effluent pH: 7, time: 2 h, inter distance between the electrodes: 1 cm and stirring speed: 100 rpm).

For Fe electrode

$$\mathrm{Fe} \rightarrow \mathrm{Fe}^{2+} + 2e^{-}$$

The nascent aluminum and iron ions are very efficient coagulants for particulates to flocculate. From the above two chemical reactions, the higher oxidation potential of Fe electrode −0.447 V than Al electrode −1.662 V, respectively. The generation of Fe coagulant is three times greater than Al which causes the higher COD removal efficiency in the iron electrode as anode, when the same electric charge is passed. All subsequent EC experiments were conducted using the Fe/Fe electrode combination system.

3.1.5. Effect of inter electrode distance

The effect of interelectrode distance between anode and cathode on the removal of percentage COD and power consumption was studied with keeping all

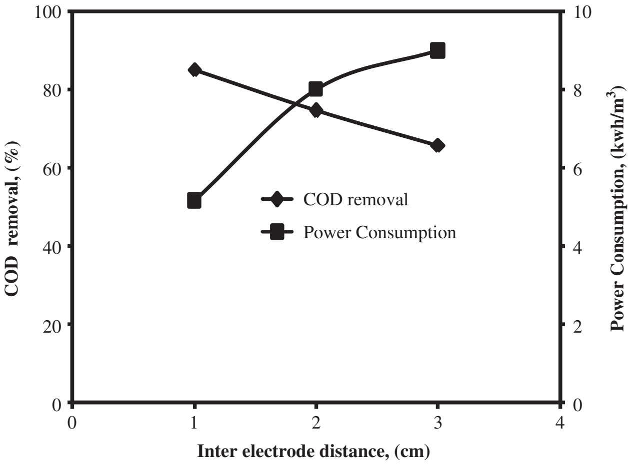

Fig. 6. Effect of inter electrode distance on percentage COD removal and power consumption (effluent COD concentration: 2,500 mg/L, current density: 0.40 A/dm^2, initial effluent pH: 7, time: 2 h, electrode combination: Fe/Fe and stirring speed: 100 rpm).

other parameters was constant, and the results were shown in Fig. 6. It can be ascertained from the figure, when interelectrode distance increased from 1 to 3 cm, the removal of percentage COD was decreased from 85 to 65.78%, and the power consumption was increased from 5.16 to 8.98 kWh/m^3, respectively.

It can attributed that increase in interelectrode distance between anode and cathode at constant current density increases IR (ohmic drop) resistances which increase the cell voltage and adversely affects the removal of percentage COD and power consumption. Therefore, the increase in IR drop is not recommended for electrochemical process for the effluent treatment in order to have acceptable power consumption as well as desired effective removal of pollutants. Hence, in order to achieve higher removal of COD in the effluent concentration, the interelectrode distance between anode and cathode should be minimized [29–34].

3.2. *Extraction of lignin from the paper and pulp mill effluent*

Following centrifugation and drying of the cake in the oven at −60˚C, amount of the lignin extracted was weighed and shown in Table 3.

3.3. *Characterization and analysis of the lignin using FT-IR*

The following Fig. 7(A)–(C) show FT-IR results for the lignin extracted from the paper and pulp mill effluent, at different pH 3, 5, and 7 values. The corresponding assignments of FT-IR spectra of the ionic liquid-soluble lignin are given in Table 3. Even though the spectra contain numerous absorption bands, not all can be designated explicitly to structural groups of lignin, and the region from the 4,000 to 3,500 cm^{-1} did not show any useful information of isolated lignin.

Table 3
Yield of Lignin corresponding to pH of effluent

pH	Product yield (g/L)
3	60.36
5	56.98
7	53.63
9	40.70
11	27.76

As can be seen from Table 4, the region 3,500–900 cm^{-1} was far more informative; however, the hydroxyl, carbonyl, methoxyl, and carboxyl groups are the most important chemical functional groups in lignin, and sufficiently, they can be used for identification of lignin [35]. Lignin extracted from kraft paper mill effluent constitutes syringyl monolignol groups, the same could be confirmed by the methoxyl groups at the side chain of lignin at characteristic 2,850 cm^{-1} band. Also, the aromatic carbon stretching indicates the polymerization of lignin monomers exhibited by the wave number bands at 869 and 1,448 cm^{-1}.

The FT-IR spectra of the lignin samples possess strong intensity band at 3,420 cm^{-1} that attributed to the hydroxyl groups, methoxyl groups are showed by medium intensity absorption bands at 2,860–2,850 cm^{-1}, and carbonyl groups are assigned at 1,740 cm^{-1} stretching frequencies with strong intensity. The strong intensity bands in the range of 1,615 cm^{-1} could be assigned to the C–C bonds group of the aromatic skeleton compounds present in the isolated lignin. The range of bands for rings conjugated with carbonyl group is found at 1,500–1,400 cm^{-1}. The medium intensity band at 1,448 cm^{-1} is attributed to the vibration of the C–H deformations and aromatic ring group. A noticeable absorption band center observed at 2,850 and 1,216 cm^{-1} reveals that the rubber wood lignin is composed of high syringyl and guaiacyl units congruent with hardwood lignin.

The medium intensity at 1,216 cm^{-1} band is also associated with aromatic phenyl C–O stretching. The band centered at 869 cm^{-1} confirmed the aromatic C–H in-plain deformation appears from the syringyl units. The intensity at 852–837 cm^{-1} resulted from aromatic group C–H out-of-plain deformation vibration as confirmed previously. Apart from that, FT-IR revealed that the absence of absorption peak of aliphatic C–H (2,925 cm^{-1}), celluloses (1,317–1,315 cm^{-1}),

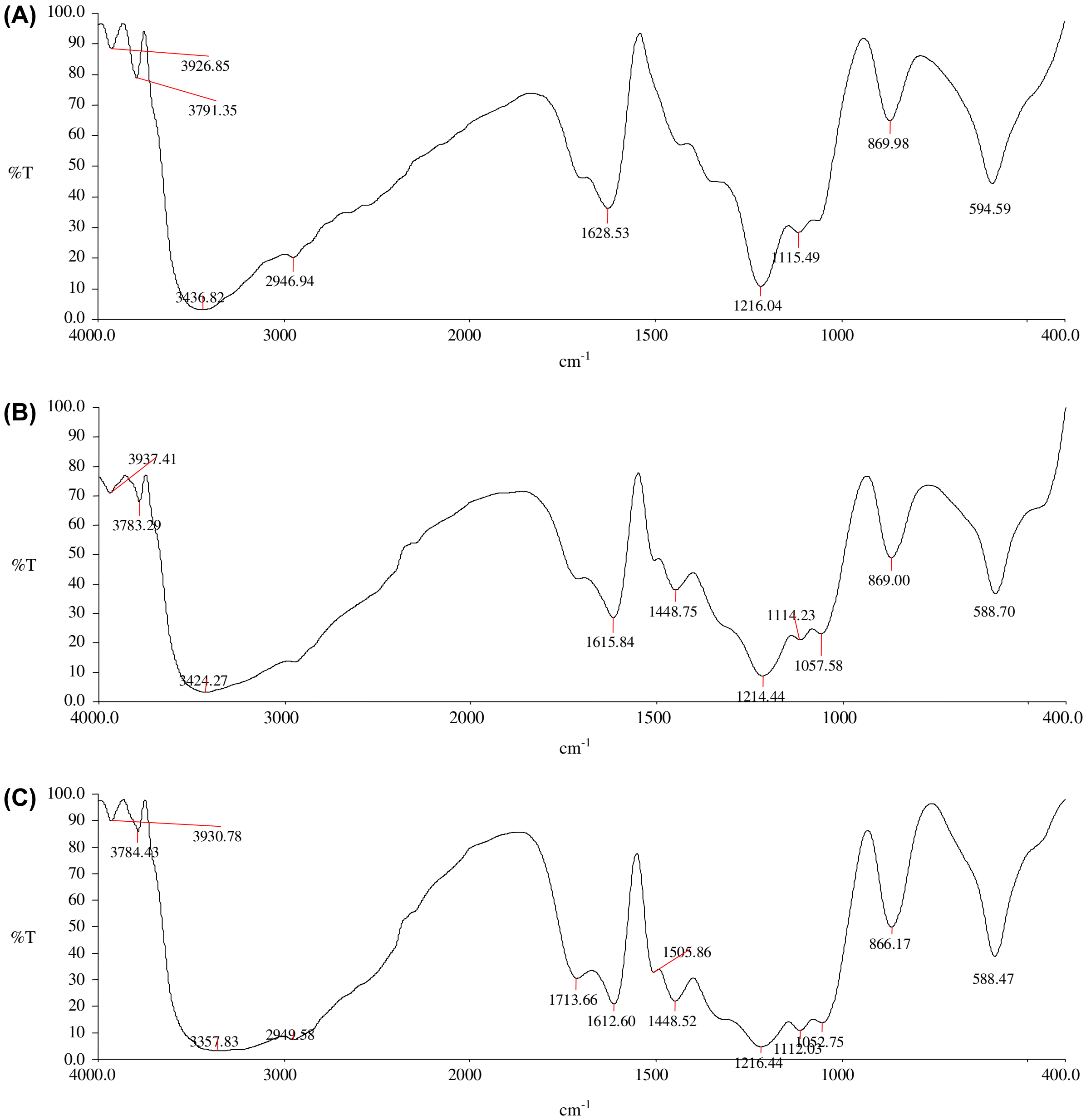

Fig. 7. FT-IR results for extracted lignin at pH (A) 3, (B) 5, and (C) 7 during EC process.

Table 4
Assignment of IR bands in the spectra

Wave number (cm^{-1})	Functional group
869	Aromatic C–H out-of-plain deformation
1,216	Aromatic phenyl C–O stretching
1,448	C–H deformations and aromatic ring skeleton vibrations
1,615	C–C bounds of aromatic ring skeleton vibrations
1,740	Carbonyl stretching
2,850	Methoxyl group
3,420	Hydroxyl vibration

hemicelluloses, and silicates (1,030–620 cm^{-1}), these non-lignin materials are mainly responsible for contamination of isolated lignin, so it clear demonstrating that the isolated lignin nearly pure.

4. Conclusion

On the basis of the experimental studies conducted, it has been observed that EC using Fe electrodes is an efficient and inexpensive alternative for the treatment of paper mill effluent. Efficacy of the process and influence of operating parameters such as current density, initial effluent pH, initial COD concentration, electrode combination, and interelectrode distance of effluent have been investigated.

It has been observed that the percentage removal of COD and power consumption increased significantly with increasing current density at 0.5 A/dm^2. At neutral initial effluent pH condition could be achieved higher pollutant removal with required power consumption. In addition, expected trend is found with initial COD concentration of effluent as well. The percentage COD removal decreased from 98 to 52% with increased COD concentration from 1,000 to 5,000 ppm and also the Fe/Fe electrode combination with minimum interelectrode distance between the anode and cathode having high pollutant removal.

Furthermore, lignin, which is a main source of color and COD in black liquor of the pulp and paper industrial effluent, has been extracted by precipitation using acid. The FT-IR analysis of the extracted lignin has revealed the likely presence of hydroxyl groups, methoxyl groups, carbonyl groups, and the absence of absorption peak of aliphatic C–H, celluloses, hemicelluloses, and silicates, thereby indicating that the isolated lignin is nearly pure.

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