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Remediation of Pulp and Paper Industry Effluent Using Electrocoagulation Process

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Abstract

Electrocoagulation of pulp and paper industry effluent with SS-304 electrode has been carried out under varying process variable such as pH, current density, time and dose of electrolyte to find out the optimum conditions. Maximum reduction efficiency of Chemical Oxygen Demand (COD) 82% and color more than 99% from pulp and paper industry wastewater at the following conditions pH = 7, current density = 24.80 mA/cm^2 time = 40 min and dose of electrolytes = 1.0 g/L. Moreover, effects of electrolytes dosage on electricity consumption were observed and found to be that NaCl is better in comparison of Na_2SO_4 in respect of lower down the electricity consumption. But application of NaCl causes the formation of hazardous compounds as secondary pollutants within treated water. Therefore, Na_2SO_4 could be a potent replacement of NaCl to enhance the conductivity of paper industry effluent treated by EC process. The treated water has been compared with standard of Central Pollution control board (CPCB) and World Health organization, and found to be suitable for the reuse in irrigation.

Keywords

Pulp Paper Industry, Electrocoagulation Treatment, Wastewater, Reuse, Chemical Oxygen Demand, Color

1. Introduction

The manufacturer of paper fundamentally relies upon the natural resources such as water, forest, agro-product, and fossil fuels (energy) for the production of paper [1]. The huge amount of fresh water is consumed coarse of Pulp and paper making process. These processes are preparation of wood, pulping, washing of pulp and bleaching which releases large volume of effluent along with high organic load, COD, Biological Oxygen Demand (BOD), colorant and lower biodegradability index. The paper industry effluent is a complicated blend of more than 250 organic and 700 inorganic chemicals [2]. Additionally, highly toxic chemicals which are resistant to biodegradation and listed as prime contaminates by United States Environmental Protection Agency (USEPA) which are related to severe health concern, generates mainly reaction between lignin and chorine/chlorine based chemical still being used for the bleaching of pulp [3]. Chlorinated phenolic compounds such as chlorophenolics, chloro-hydrocarbon, and fatty acid are also present in wastewater. Some of them *i.e.* chlorophenolics, dioxin and furan show the higher toxicity. When this effluent is released without appropriate treatment, it influences the negative effect on receiving water bodies involving thermal impacts, low Dissolved Oxygen (DO), poor sunlight penetration, scum formation, slime growth, and harming the aesthetic and scenic beauty of environment and disturbed the overall ecosystem. Some investigator reported the toxic effect on fish present and growing in the water bodies containing paper industry effluent. The Indian paper industry used mainly two-step treatment at their treatment plants such as primary (sedimentation, floatation) and secondary (activated sludge, anaerobic or aerobic process). These methods are failed to meet the requirements of complete degradation of bio-refractory compounds, COD and color including some drawback such as high treatment cost and generating high volume of sludge, which consequently may be responsible for secondary pollution. Hence, there is an urgent need to stop the formation or to impose suitable measures for the degradation of these dreadful chemical by modification in plant. Meanwhile an effective treatment facility is becoming a necessity to remove these pollutants from wastewater [3] [4] [5] [6].

Nowadays electrocoagulation (EC) method has intensively attracted the researchers from around the world due to its unique feature in comparison to the chemical and biological treatment facility. EC process exhibits environmental compatibility, proper setup, shorter reaction time, meager chemical requirement along with negligible generation of sludge [7]. The main acting reagents in this process are electrons which are clean reagent [8]. Recently, EC method has been considered as an advanced oxidation process and successfully applied for treatment of the different types of industry effluent including organics matter, phenolic compounds, potato chips manufacturing unit, methylparathion from textile unit, colorants from dye solutions, oil suspensions, and fluorides [9]-[15]. When the direct current is supplied between anode and cathode, the metal plates dissipate and release the positive hydroxide ions at desired pH [16]. In the present investigation, stainless steel (SS)-304 has been used as electrode material. When oxidation occurred at Fe anode, it may produces iron hydroxide, $Fe(OH)_n$, where $n = 2$ or 3. The mechanisms for the production of $Fe(OH)_n$ of the process is given as follows:

At anode:

$$Fe_{(s)} \rightarrow Fe^{2+}_{(aq)} - 2e^{-} \quad (1)$$

$$Fe^{2+}_{(aq)} + 2(OH)_{(aq)} \rightarrow Fe(OH)_{2(s)} \quad (2)$$

At cathode:

$$2H_2O_{(l)} + 2e^{-} \rightarrow H_{2(g)} + 2OH^{-}_{(aq)} \quad (3)$$

Overall reaction:

$$Fe_{(s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + H_{2(g)} \quad (4)$$

These $Fe(OH)_n$ metal ions behave as coagulant or destabilize the pollutants present in wastewater by charge neutralization and having a great potential to separate them by electro-flotation. In recent years, the EC process has usually been used for the separation of pollutants present in different types of water and wastewater [7] [17] [18]. As for increasing environmental awareness and strictness in polices, force to adopt integrative management practice for wastewater treatment. On the other hand, research related to wastewater treatment and reuse is poorly reported. As per our knowledge very hardly any study has been conducted to develop the optimum treatment conditions and suggestion of reusability of EC treated water for irrigation. The aims of this work to determine the optimum condition correspondence to the maximum COD and color removal from paper industry waste effluent. Further, find out an alternative option for the utilization of treated water in agriculture.

2. Material and Methods

2.1. Characteristics of Effluent

The wastewater has been collected after primary treatment from nearby paper industry followed by immediate storage at temperature below than 4˚C. The analysis of the samples for several parameters such as pH, BOD_3, COD TOC, color and TDS were performed as per standard method [5] and observation are given in **Table 1**. The conductivity and pH of the effluent have been performed by a digital conductivity meter and calibrated pH meter, respectively. The treatment efficiency (TE) was also determined in terms of maximum COD and color elimination percentage from effluent by the following equation;

$$\text{Treatment efficiency}(\text{TE}\%) = C_0 - C_t / C_0 \times 100 \quad (5)$$

where,

TE = Treatment efficiency (%), C_0 = Initial concentration of pollutants, C_t = concentration of pollutants after time (t).

$$\text{Total Energy consumption}\left(\text{kWh/m}^3\right) = \frac{VIt}{\text{Treated volume}\,(l)} \quad (6)$$

where, V = Volume potential of electricity (V), I = Current (ampere), t = time of treatment (h) and l is the treated volume in liter [19].

Table 1. The major characteristics of paper industry effluent.

S. No.	Parameters	Values
1	pH	7.65
2	BOD (mg/L)	176
3	COD (mg/L)	534
4	BOD/COD	0.32
5	TOC (mg/L)	209
6	Color (Pt. Co. Units)	1154
7	TDS (mg/L)	1858
8	Conductivity (mS)	3.32

2.2. Electrocoagulation Unit

The batch scale laboratory diagram of experimental assembly is depicted in Figure 1.

The electrode configuration and EC experiment details are depicted in Table 2. The EC tests have been done in a 250 ml plastic beaker at ambient temperature. Direct current power supply was used for supplying the current in reaction vessel. The stirrer has been used at 100 rpm for continuous stirring of media during the test. EC method has its own drawback as it uses vast quantity of electricity (exhaustible source). Keeping the environmental concern in mind, the experimental setup is connected to the photovoltaic cell (solar energy) to reduce the electricity

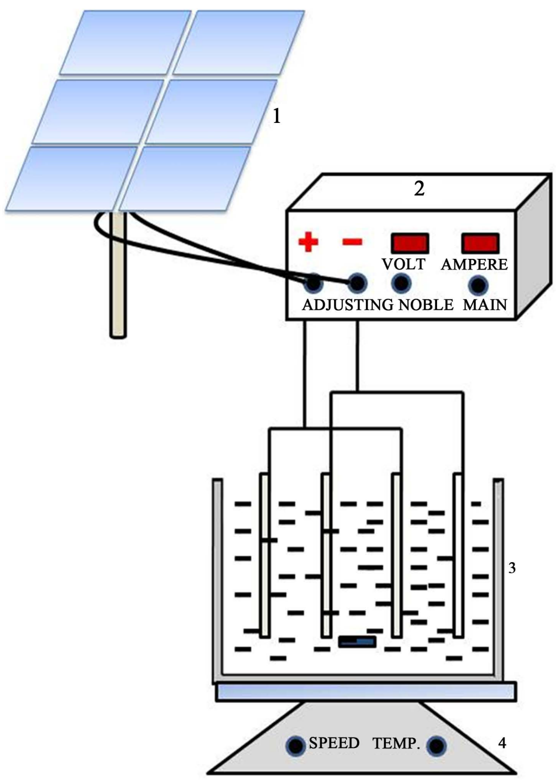

Figure 1. Laboratory scale experimental setup of electrocoagulation unit (1-Solar panel, 2-Direct current supply and 3-Treatment vessel, consists of Anode and Cathode in mono-polar mode, magnetic-bead, and 4-magnetic stirrer).

Table 2. Characteristics of electrode and treatment assembly.

S. No.	Electrode characteristics	
1	Material (anode and cathode)	SS-304
2	Shape	Rectangular
3	Size (mm)	40 × 70
4	Number	4
5	Plate arrangement	Parallel
6	Connection mode	Monopolar
7	Effective area of electrode (mm^2)	504
	Reactor characteristics	
8	Make	Plastic
9	Mode	Batch
10	Volume (ml)	250
11	Used water volume (ml)	250
12	Electrode gap (mm)	10
13	Power supply	Direct current
14	Voltage range (V)	1 - 12
15	Current range (A)	4 - 35 mA/cm^2

consumption. During the EC treatment, direct current is used so that solar energy could be quite meaningful to dealing with the treatment of water and wastewater at laboratory and industrial scale. Since India is considered to be a tropical country and having vast solar energy potential, it could be more attractive option for the usage of EC process. Moreover, Photovoltaic cells produce the direct current which is directly used in EC process without any change making it an affordable alternative for the treatment of wastewater facilitated by EC process. Earlier reports have also asserted on the successful treatment of different types of industrial effluents by EC [20] [21] [22] [23] [24].

3. Result and Discussion

3.1. Parameter Optimization

Pulp and paper industry effluent found to be intensively polluted in nature with heavy organic load in terms of COD and color. This study investigates the optimum conditions for EC on the basis of maximum COD and color removal by varying several variables such as pH (5.0 - 9.0), time (10 - 50 min), current density (4.96 - 34.72 mA/cm^2) and electrolytes dose (0.5 - 2.0 g/L).

3.2. Effect of pH

It has been extensively studied that the pH is one of the crucial parameter to govern the efficiency of EC method [25] [26] [27]. In this investigation, the effect of pH was observed for the reduction of COD, and Color from paper industry wastewater for its treatment. A sequence of experiment was conducted by varying the pH from pH 5.0 to pH 9.0, while other parameters like current density 5 mA/cm^2, time 10 min and dose of electrolyte 0.5 g/L remained constant. This is clear from **Figure 2** when the pH of media was changing from 5 - 7, the COD and Color reduction rises gradually due to the occurrence of freshly formed $Fe(OH)_n$ and maximum reduction was found to be near pH 7.0. These Fe ions possess a large surface area and higher affinity for coagulation. In addition, it was also observed that pH in basic medium causes the solubility of $Fe(OH)_3$ to increase which leads to the formation of $Fe(OH)_4$ which ineffective for the treatment of wastewater treatment [27] [28] [29] [30]. The efficiency of treatment was not observed to be good at acidic or basic medium while it is lowest at pH 5 and pH 9 respectively.

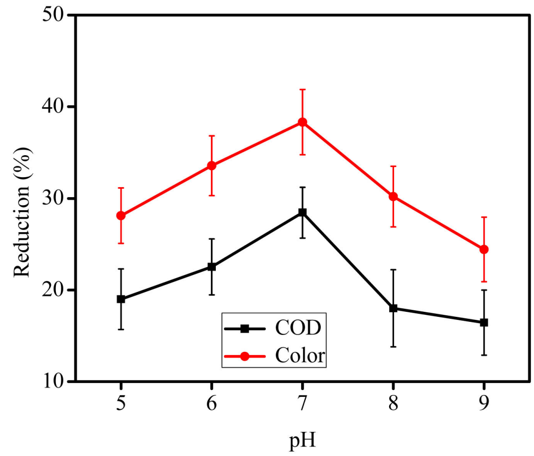

Figure 2. Effect of pH on COD and color removal.

Moreover, the EC method exhibits buffering effect which makes the EC process unique in terms of feasibility for wastewater treatment. Therefore, it is not required to maintain the pH of treated water before releasing it into water system. Additionally the treated water of either acidic or basic pH can cause several negative effects on receiving water bodies and entire ecosystem [5] [31].

3.3. Effect of Current Density

Various study described that current density is a major parameter which leads to the higher treatment efficiency and is also cost effective for the EC process [19] [29] [32] [33]. The current density was varied from 4.96 - 34.72 mA/cm^2 for reducing the COD and color content from waste effluent (**Figure 3**). It was noticed that as the current density increases, it led to greater amount of metal dissolution or anodic oxidation which in turn leads to high precipitation of hydroxy cationic complexes. It is obvious that applied current density enhance the rate of bubble production along with the rate of coagulant (flocs formation) which is beneficial for the treatment efficiency of the EC.

Besides, it is also reported that bubbles density increases and their size decrease

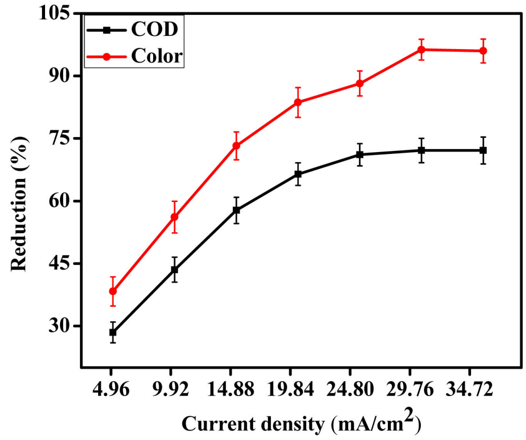

Figure 3. Effect of current density on COD and color removal.

by means of rising current density ensuing a larger upwards flux and more rapid removal of pollutants and sludge flotation [33] [34]. In this study it was observed that the removal efficiency of pollutants and current density are directly proportional to each other.

3.4. Effect of Treatment Time

The effect of treatment period was investigated to varying time intervals (10 - 50 min), pH, current density, dose of electrolyte fixed at 7, 24.80 mA/cm^2 and 0.5 g/L, respectively to find out the reduction in COD and color in present effluent sample. At the beginning of the run, the solution was transparent which progressively becomes dark and after about 30 min it turns greenish as the reaction progress due to formation of $Fe(OH)_{n(s)}$. After about 10 min a significant reduction in color was observed (**Figure 4**) and at the end of the treatment (~40 min) reduction in color value reached up to 97% (**Figure 4**). The color of Fe ion disappears slowly with the progression of time since the produced Fe ions mostly coagulant with the pollutant species and form settleable sludge [35] [36] [37]. Same pattern was observed in the case of COD reduction, as it was observed that an increasing in the time leads to increase in the removal efficiency of COD. A

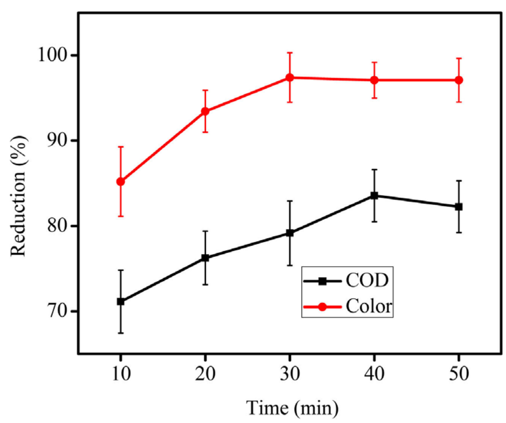

Figure 4. Effect of time on COD and color removal.

removal of 81% in COD value was observed in 40 min time and after that the small changes occurred in value of COD and finally plot become constant.

Thereafter, no reduction was observed in the COD and color found to be in a nearly steady state. Therefore, 40 min is taken as the optimum time for the degradation of the organic matter in the present effluent sample [37].

3.5. Effect of the Supporting Electrolyte on Process Efficiency and Electricity Consumption

The conductivity of solution plays a key role to boost the process efficiency and reduce the current density. Conductivity maintains the lower potential flowing in circuit during the treatment and reduced electricity energy consumption (EEC) [38]. The heterogeneous ions transferred in the EC method due to the conductivity of the effluent. In this treatment two electrolytes have been used which are Na_2SO_4 and NaCl.

The effect of variable concentrations of electrolytes (0.5 - 2.0 g/L) was observed on the removal of chemical oxygen demand and color and the EEC consumption. EC process was performed using current density of 24.80 mA/cm^2, time 40 min and pH 7. As shown in **Figure 5**, when the concentration of both Na_2SO_4 and NaCl from 0.5 to 1 g/L the increment in removal efficiency was observed, from 79 - 84. This may be due to the rapid movement of ions of electrolytes. Moreover, when the supporting electrolyte concentration is increased to 2 g/L, no further improvement is found in removal efficiency which may be due to an increasing of the passivation layer [27] [39].

The existence of the sustaining electrolyte also cut down the EEC, as revealed in **Figure 6**. This may be due to the increase of the conductance of solution, and leads the countable effect in EEC from 28 kWh/m^3 to 16.5 kWh/m^3 using 1 g/L supporting electrolyte (**Figure 6**). Consequently, a value of 1 g/L concentration of the electrolyte was preferred to acquire high removal efficiency of treatment, as well as to reduce EEC. During the treatment EEC of the wastewater was also reliant on process time. The EEC found to be directly proportional with increasing process time and linearly increases from 0.848 to 16.50 kWh/m^3 with an increasing process time from 10 to 40 min at the 24.80 mA/cm^2 current density and 1 g/L Na_2SO_4. Additionally, a highest COD reduction of near about 82% was observed after 40 min, whereas reduction in COD at 60 min was about 80%. The EEC at 50 min was found to be higher in comparison of 40 min (**Figure 6**). A similar result was obtained for 1 g/L NaCl. A removal efficiency of 82% was obtained using Na_2SO_4 at 16.5 kWh/m^3 EEC, while same removal efficiency was found using 16.17 kWh/m^3 EEC when NaCl used as electrolyte. NaCl was observed quite effective in respect of EEC consumption in comparison to $NaSO_4$ [17] [27]. Additionally, If NaCl used as electrolyte during the treatment of paper industry effluent, it may be quite harmful due to the presence of Cl^- ions. Additionally, once the chloride ions added in the solution, they produced Cl_2 and OCl^- at anode. This OCl^- take part in oxidation reaction and remove organic matter. These ions may reacts with lignin and phenols already present in wastewater,

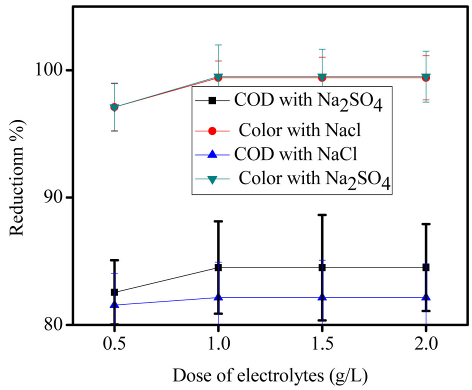

Figure 5. Effect of dose of electrolyte on COD and color removal.

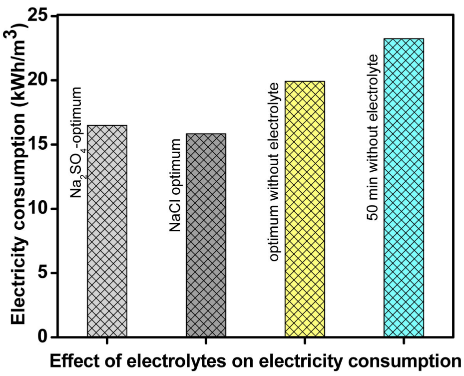

Figure 6. Effect of dose of electrolyte on electricity consumption.

and produced water soluble electrolignin, easily oxidizable chlorolignin and chlorophenolics compounds. Earlier researcher suggested that these compounds caused higher toxicity and disturbances in aquatic system [7] [40] [41]. Thus, Na_2SO_4 could be more favorable option for electrolyte for the treatment of pulp and paper industry wastewater using EC methods.

3.6. Kinetics Study

The kinetics of electro-coagulation was studied in order to assist the rate of reaction during the process of oxidation. Paper industry wastewater contains a mixture of various compounds with differential reactivity. Therefore, it is quite difficult to perform a detailed analysis of individual compounds. To study such a complicated process, the rate of reaction was studied in terms of a combined parameter (*i.e.* COD).

The experimental data observed with time-dependent COD removal in the presence of Na_2SO_4 as electrolyte modeled by the assumption of first-order kinetics where the progressive disappearance of COD can be presented as the percentage COD removal is proportional to the pollutant concentration and the numbers of hydroxides [flocks] generated (**Figure 7**). The rate equation can be written as [42]:

$$\frac{-\mathrm{d}[\mathrm{COD}]}{\mathrm{d}t} = k[\mathrm{COD}][\mathrm{Fe(OH)}_3] \quad (7)$$

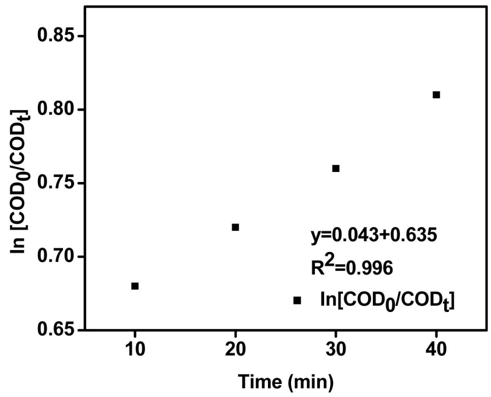

Figure 7. Linear fitting of ln[CODo/COD_t] as a function of reaction time.

By the integration of equation (9) yields

$$\log\frac{[COD_t]}{[COD_0]}=-kt \tag{8}$$

where COD_0 refers the initial effluent COD and COD_t refers the effluent COD at time *t*.

The reaction rate constant *k*, can be estimated from the plot log[COD_t/COD_0] versus electrolysis time. The experimental data fitted well to first order kinetics, as a straight line with an R^2 value of 0.996 been critically examined.

3.7. Reuse Option for Treated Water

Water shortage is a fundamental problem at present in various parts of world which is badly influencing a huge portion of the world population. With this reality, farming emerges as one of the high water demands, it consuming about 70% of worldwide freshwater. Besides, 28% - 56% of the worldwide irrigated cropland is situated in regions under high (40% to 80%) or very high (>80%) water pressure, in light of the proportion of water withdrawal over accessible water [43]. In this sense, water recovered from municipal wastewater has turned out to be one of the major and more affordable non-regular water hotspots for farming which is, with about 20 out of 200 million Ha (hectare) of irrigated land around the world, the main recycled water consumer and one of the less expensive in which its utilization demonstrates its genuine advantages. The irrigation of agricultural land by using reclaimed water having some advantages such as reduce the fresh water consumption and pressure over the fresh water sources, added high nutrient value to soil which diminish the uses of synthetic fertilizer and enhance the yields of crops in comparison of freshwater irrigated crops. On the other hand, mismanagement in wastewater reclamation may cause negative effect on human health along with environment. There are most possible concern is presence of pathogens which may enter in the food chain and can cause several diseases to flora and fauna. Additionally, land and crops might be affected by expanding salinity. (Phytotoxic components can influence development of products decreasing yields and the structure of soils may result in harm because of high sodicity levels.) Untreated wastewater discharges into the streams where it is diverted by subsistence agriculturists to little plots of vegetables and greens serving of mixed greens crops created for adjacent urban markets [44]. These primary vegetables are carrots, lettuce, cabbage and others which are smoothly devoured crude as plate of mixed greens and green vegetables. The general prosperity risks of using such dirtied streams for water framework are undeniable. Treated gushing can be used for horticulture framework under controlled conditions to restrain the trading of pathogenic and hurtful contaminants into the cultivating, soils, surface, and groundwater. The issues of lack of water and transfer wastewater in dry zones can be decreased by the use of treated water in agribusiness fields. Especially for less prolific soil, this might be fundamental wellspring of enhancements for harvest creation. Usage of treated water to cropland and forested region is an appealing option for exchange since it can improve the physical properties and the enhancement substance of soils [45]. Water system with treated water gives, supplement, for example, N and P and adding natural issue to the dirt, yet there is a dread about the gathering of perhaps unsafe segments, for instance compact disc, Cr, Cu, Pb, Fe, Al, Mn, Zn, phenols, chlorophenol and their subordinates from paper plants wastewater [46]. In this study treated water was compared to the standard of CPCB and WHO provided for the irrigation in agriculture.

According to **Table 3**, all residual operating parameters were in the prescribed range of World Health Organization (WHO) and CPCB standard that ensure the reusability of treated water for irrigation purposes. This EC process and its applications are environmentally sustainable and economically viable.

Table 3. Comparison of studied treated water quality with CPCB and WHO wastewater reuse standards for irrigation in agriculture land [47].

Characteristics	Treated water	PL* (CPCB)	PL* (WHO)
pH	7.25	5.5 - 9.0	6 - 9
BOD (mg/ L)	52	100	200
COD (mg/ L)	91	NA	500
TDS (mg/ L)	1059	NA	1500
Color (Pt-Co. Units)	6.93	NA	NA
Conductivity (mS)	2.83	NA	NA

*PL = Permissible Limit.

4. Conclusion

The EC method provides inspiring results from the treatment of pulp and paper industry wastewater. The investigation shows that the EC method is a versatile process and has the potent removal efficiency towards the BOD (Biochemical Oxygen Demand), COD (Chemical Oxygen Demand), Color, TOC (Total Organic Carbon) and TDS (Total dissolved solid) from pulp and paper industry effluent. The overall reduction in COD, Color, TOC and TDS was 82.00%, 99.4%, 79% and 43% respectively; biodegradability ratio enhanced up to 0.52 at pH = 7.00, current density = 25.20, time = 40 min and a dose of electrolyte 1 g/L with 100 rpm stirring speed. The analysis was repeated in triplicate and result showed the good repeatability in the error range of 2% - 5%. All the physiochemical parameters of EC treated water were found below the permissible limit prescribed by WHO (World Health Organization) and CPCB (Central pollution Control Board); thus, can be used as an alternative to reduce the fresh water input in agriculture fields. The pH of the treated water after EC treatment found to be within permissible limit of environmental standard, it is obtained near neutral pH. It is meaningful feature of EC method and not being needed neutralization process in comparison to conventional treatment.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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