Treatment of paper-recycling wastewater by electrocoagulation using aluminum and iron electrodes

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Received: 22 April 2018 / Accepted: 5 September 2018 / Published online: 15 September 2018
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Abstract

Treatment of industrial wastewater by electrocoagulation (EC) is one of the most efficient methods to remove pollutants. Paper-recycling wastewater is a complex mixture containing toxic and recalcitrant substances, indicating complexity and difficulty of its treatment. The aim of the present study was to assess the effectiveness of paper-recycling wastewater treatment by EC process using aluminum (Al) and iron (Fe) plate electrodes. Removal of chemical oxygen demand (COD), total suspended solids (TSS), color and ammonia from paper-recycling mill effluent was evaluated at various electrolysis times (10–60 min), voltage (4–13 V) and pH (3.5–11). The optimum process conditions for the maximum removal of COD, TSS, color and ammonia from paper-recycling industry wastewater have been found to be pH value of 7, treatment time of 60 min and voltage of 10 V. Under optimum operating conditions, the removal capacities of COD, TSS, color and ammonia were 79.5%, 83.4%, 98.5% and 85.3%, respectively. It can be concluded that EC could be considered as an effective alternative for treatment of paper-recycling wastewater.

Keywords Paper-recycling wastewater · Electrocoagulation · Aluminum electrode · Iron electrode · Power consumption

Introduction

The pulp and paper industry is considered as a highly water-intensive one with significant amounts of wastewater discharge. More than 250 chemicals are produced at different stages of pulp and paper production which are found in the wastewater. Discharge of untreated pollutants into the environment can lead to various environmental problems due to their accumulation in recipient water and soil. This type of wastewater often includes a high level of COD, BOD, wood debris and other contaminants. However, wastewater composition varies according to the type of processes used in the plants [1–4].

Several treatment technologies have been applied in order to prevent the harmful effects of effluents on the environment, and therefore, the design and efficiency of wastewater treatment techniques varies among mills [5–7]. The main treatment processes in pulp and paper industries consist of primary clarification (sedimentation or flotation), secondary treatment (activated sludge process or anaerobic digestion) and tertiary processes (membrane processes) [7]. Activated sludge plant has been the most common wastewater treatment process for the removal of organics. However, this process suffers several drawbacks, such as production of sludge with very variable settlement properties, sensitivity to shock loading and toxicity and also the limitation of capacity to remove non-biodegradable substances [8].

However, one of the most interesting and effective processes for treating some pollutants in wastewater is the electrochemical processes. Electrochemical processes can be applied in various configurations such as electro-oxidation, electro-precipitation, electrocoagulation, electro-deposition and electro-Fenton [9, 10]. The electrochemical processes for treatment of water and wastewater are considered advanced oxidation processes which have been widely applied as attractive and suitable

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methods by various benefits including environmental compatibility, adaptability, energy efficiency, selectivity and cost effectiveness [11, 12].

Electrocoagulation as an electrochemical process, which uses an electrical current, has been proved to be an efficient method for the treatment of industrial wastewater [13]. This method allows wastewater to electrochemically oxidize or reduce the organic contaminants to non-hazardous inorganic substances [14] and is specified by simple equipments, environmental compatibility, safety, selective capacity, easy functionality, short resistance time, negligible equipment for adding chemicals, high efficiency for water purification, stability of sludge and the decreased amount of sludge [15–18].

It is generally accepted that the electrocoagulation process includes three stages: 1) formation of coagulants by electrolytic oxidation of the electrode, 2) the destabilization of contaminants, particulate suspension and breaking of emulsions, 3) aggregation of the destabilized particles to form flocs [19]. In this process, coagulants are produced by the electro dissolution of a sacrificial anode, which is usually made of Al or Fe. Generation of metal ions occurs at the anode, and hydrogen gas is released from the cathode. The hydrogen gas also helps to float the flocculated particles on the surface of water [20–22].

Anode and cathode reactions for Al electrodes are as follows (Eqs. 1, 2):

Anode and cathode reactions for Fe electrodes are as follows (Eq. 3–5):

Fe^{3+} and Al^{3+} ions generated by electrochemical oxidation in EC process may form monomeric and polymeric irons and aluminum species, which finally transform into $\text{Fe}(\text{OH})_3(\text{s})$ and $\text{Al}(\text{OH})_3(\text{s})$ depending on the pH of the aqueous medium (Eqs. 6, 7) [21–23].

In other words, the produced Al^{3+} , Fe^{3+} and Fe^{2+} ions react with hydroxyl to form amorphous hydroxide flocs. Accordingly, the mechanism of removing pollutants with both electrodes is associated with formation of $\text{Fe}(\text{OH})_3(\text{s})$, $\text{Al}(\text{OH})_3(\text{s})$, monomeric/polymeric iron and aluminum species [21]. Furthermore, freshly formed amorphous $\text{Al}(\text{OH})_3(\text{s})$ and $\text{Fe}(\text{OH})_3(\text{s})$ sweep flocs in the EC process have large surface areas which are helpful for a rapid adsorption of soluble organic compounds and trapping of colloids [22].

EC process was assessed successfully for treating municipal wastewater [24], landfill leachate [25], textile wastewater [26, 27], heavy metal contaminated water and wastewater [28–30], vegetable oil refinery wastewater [31], dairy wastewater [32], dye contaminated wastewater [33] and olive oil mill wastewater [34]. In addition, some studies have reported treatment of pulp and paper mill wastewater using electrocoagulation process [35–38]. However, there has been no reports on treatment of paper-recycling plant wastewater by electrocoagulation using combination of Al and Fe electrodes.

Operating cost analysis was conducted for removal of pollutants from wastewater of lather finishing industrial process using EC. The results showed the EC was cheaper compared with the conventional methods. The operational cost of EC was found to be US \$ 1.7 per cubic meter of the treated effluent as compared to the cost of US \$ 3.5 per cubic meter for conventional methods [39]. It has been reported that the operating cost of chemical coagulation is 3.2 times as high as that of electrocoagulation for the treatment of textile wastewater [40].

In the present study, we have conducted an experimental investigation of treating paper-recycling mill effluent using the electrocoagulation method. The efficiency of electrocoagulation in removing COD, TSS, color and ammonia from paper-recycling mill effluent was explored. The effect of several experimental parameters such as pH, voltage and electrolysis time on the removal efficiency was investigated to determine the optimum operational conditions.

Materials and methods

Characterization of the paper-recycling mill effluents

The paper-recycling wastewater samples were taken from Kahrizak paper mill located approximately 10 km far from Tehran (Tehran province, Iran). All samples were collected manually in 20-l polyethylene containers and transferred to the laboratory, and they were also stored at 4 °C to reduce

biodegradation. The main characteristics of the raw wastewater are presented in Table 1.

Electrocoagulation set-up and measurements

EC process has been evaluated for the reduction of a wide range of pollutants using batch or continuous mode of operation. Comparably, a batch reactor's dynamic nature allows for studying the range of operating conditions and is better suited for laboratory and pilot plant scale applications, while the continuous system is more suited to industrial processes with large effluent volumes [11, 41]. As shown in Fig. 1, electrocoagulation experiment was carried out in a batch mode reactor made of Plexiglas with dimensions of 20 cm × 10 cm × 17 cm.

In the electrocoagulation process, the electrode type is an important parameter and has a significant effect on removal efficiency. Iron and aluminum plates were used as electrodes in the electrocoagulation process because of their non-toxic nature, being cheap, easy accessibility and simple production [42, 43]. It has been found when COD, color and phenol removal are important, application of Al and Fe electrodes combination has higher efficiency in paper mill wastewater treatment [44]. In our study, there were two iron electrodes between two aluminum electrodes arranged in monopolar configuration and effective area of each electrode was 14 cm by 8 cm. Samples from the electrocoagulation unit were allowed to settle for 30 min in a beaker before every analysis. The electrodes were washed with HCl solution (15%W/V) before each run. Following each run, the electrodes were washed with distilled water, then dried and used again.

A digital power supply (Zhaoxin RXN-605D), which transform alternating current to direct current, was used as an energy source in EC process at a constant temperature of 23–25 °C and 100 rpm stirring speed. The maximum output voltage and current of the power supply were 60 V and 5 A, respectively.

Table 1 Characteristics of paper-recycling mill wastewater

Characteristics	Paper-recycling mill wastewater
Color (Pt.Co ^a)	8000
Temperature (°C)	26–31
pH	7.2
COD (mg l ⁻¹)	900
TSS (mg l ⁻¹)	4120
TDS (mg l ⁻¹)	7000
NH ₃ (mg l ⁻¹)	4.5
Turbidity (NTU ^b)	2440

^a Platinum cobalt unit

^b Nephelometric turbidity unit

Fig. 1 Bench-scale EC reactor with monopolar electrodes in parallel connection

The measurement of COD, TSS, color and ammonia before and after electrocoagulation was carried out according to the Standard Methods (American Public Health Association standards) [45] to calculate the removal efficiency of pollutants. Concentrations of COD and ammonia were analyzed colorimetrically with a spectrophotometer (Hach DR 5000, USA). The color and turbidity were measured by a Hach DR-2000 spectrophotometer.

Results and discussion

Effect of electrolysis time

Electrolysis time influences the treatment efficiency of the EC process. During electrolysis, anodic electro-dissolution leads to the release of coagulant species. Besides, removal efficiency of pollutants depends directly on the concentration of metal ions produced on the electrodes. When the electrolysis time increases, the concentration of metal ions and their hydroxide flocs increases [46, 47]. Theoretically, based on Faraday's law, the amount of released coagulants from aluminum electrodes tends to increase with electrolysis time [48]. Therefore, the graph of pollutants removal versus retention time demonstrate an increasing trend [49].

The COD reduction percentage, as an indicator of the pollutants removal, increased with increasing the electrolysis time. Because the Fe(OH)₃ flocs are generated by applying current in the electrodes, and therefore, they adsorb the organic molecules of the effluent [50].

In this study, investigations regarding the effect of electrolysis time on the removal of pollutants were conducted. Figure 2 illustrates the efficiencies of COD, TSS, color and ammonia removal at different electrolysis times (10, 20, 30, 40, 50, and 60 min) with an initial pH of 7.2 and a constant cell voltage of 4.0 V. As it is obvious, by increasing electrolysis time, the

Fig. 2 Effect of treatment time on COD, TSS, color and ammonia removal (initial pH value of 7.2 and voltage value of 4 V)

efficiency of pollutants removal increased. In the first half of the operating time, 81% and 78% of color and ammonia removal, respectively, were achieved. The generation of Fe^{3+} at the anode is the reason for particle aggregation-polymerization. Depending on the initial pH of the wastewater, this ion may form ferric hydroxo-complexes and polymeric species such as $Fe(H_2O)_6^{3+}$, $Fe(H_2O)_5OH^{2+}$, $Fe(H_2O)_4(OH)_2^+$, $Fe_2(H_2O)_8(OH)_4^{4+}$ and $Fe_2(H_2O)_6(OH)_4^{2+}$. The amount and variety of these species that are key factors in removing contaminants depend on electrolysis time [50, 51].

The removal of TSS, COD and ammonia during the treatment time indicates an approximate steadiness after 50 min, but there is a relatively considerable color removal between 50 and 60 min. Therefore, 60 min of operation time was applied for further studies.

Effect of voltage

Current density and voltage are important factors in the electrocoagulation process for removal of contaminants [34, 49]. The voltage of electrolysis is related to the distance of electrodes, conductivity, electrode surface state and current density [52].

As the voltage increases, the length of EC process decreases. Due to a sufficient voltage through the solution, the metal ions generated by dissolution of the sacrificial electrode are hydrolyzed to form a series of metallic hydroxide species. Therefore, these species neutralize the electrostatic charges on the dispersed particles to reduce the electrostatic inter-particle repulsion to the value small enough for the van der Waals attraction to predominate and accordingly facilitate agglomeration [21, 53].

Fig. 3 Effect of voltage on COD, TSS, color and ammonia removal (initial pH value of 7.2 and operation time value of 60 min)

Fig. 4 Effect of pH on COD, TSS, color and ammonia removal (voltage value of 10 V and operation time value of 60 min)

In our experiments, the electrolysis time was set at 60 min and initial pH of the wastewater was 7.2. The effect of different voltages (4, 6.5, 10, 13 V) on the removal of COD, TSS, color and ammonia was investigated. As can be seen in Fig. 3, at 4 V, more than 70% of pollutants were removed and the efficiency of color removal was greater than 90%. When the applied voltage was raised to 10 V, the removal efficiency of COD, TSS and color rose. Because a larger amount of Fe^{2+} was produced via anodic metal dissolution, which is able to react with dissolved oxygen of the wastewater according to Eq. (8), causing coagulation and more bubbles formed at the cathode, leading to flotation [50].

Between 10 and 14 V, a decline in TSS removal was observed, and therefore, the optimal voltage for the

process was 10 V. Results at this point were 78.2%, 83.1%, 95.6%, and 85.5% for removal of COD, TSS, color and ammonia, respectively.

Effect of initial pH

The pH is another important factor that has a notable influence on the performance of electrocoagulation process [44, 54, 55]. As shown in Fig. 4, the removal efficiency of COD, TSS, color and ammonia was analyzed at different pH values. These experiments were conducted in 60 min and 10 V, which were the optimum conditions achieved in the previous sections. In order to adjust pH, the H_2SO_4 and NaOH solutions were applied.

The results showed that when pH of the wastewater was between 5 and 10, removal efficiency of pollutants was optimal, which is close to the optimal pH range for $\text{Al}(\text{OH})_3(\text{s})$

Fig. 5 The pH variation of the wastewater before and after treatment (voltage value of 10 v and treatment time of 60 min)

formation. The flocs of $\text{Al}(\text{OH})_3(\text{s})$ have large surface areas, which are useful for a rapid adsorption of soluble organic compounds and also trapping of colloidal particles [46, 56]. pH of 7 was selected as the optimum value, considering the fact that the initial pH of raw wastewater was 7.2 (no need to add chemicals), and efficiency of EC at this point is acceptable. At pH value of greater than 10, the removal efficiency of color and TSS were reduced because at high pH solution, the dominant species is $\text{Al}(\text{OH})_4^-$, which obviously does not coagulate the pollutants [57].

Generally, the pH of the medium changes during the EC process and this change depends on the type of electrode material and on the initial pH [49, 58, 59]. According to Fig. 5, at pH values of less than 7, pH of wastewater increases after treatment by electrocoagulation. This increase was assigned to hydrogen evolution at cathodes [60]. However, this was contested by Chen et al. [61] who described this increase by the release of CO_2 from wastewater owing to H_2 bubble disturbance. In other words, at low pH, CO_2 is oversaturated in wastewater and can be released during H_2 evolution, causing an increasing of pH. In alkaline medium (pH > 8), the final pH does not change very much, and a slight drop was obtained. This result was in accordance with other reports and suggests that EC can act as pH buffer because of the balance between the production and the consumption of OH^- ion [34, 55, 62].

Power consumption

Cost evaluation plays an important role in industrial wastewater treatment because the wastewater treatment technique should be cost-effective. The costs involved in EC mainly include the energy consumption, the electrode consumption and the addition of any external chemical for increasing the efficiency of EC. In general, electrical energy consumption is a very important economical parameter in the electrocoagulation process and can be calculated using the following equation [11, 63, 64]:

$$C_{\text{energy}} = \frac{(U \times I \times t)}{V} \quad (9)$$

where C_{energy} is the consumed electrical energy (kWh/m^3), U is the cell voltage (V), I is the current in ampere (A), t is the treatment time (h) and V is the volume of the solution (m^3). In this study, the reactor-specific energy consumption under optimum conditions (pH 7, 10 V, 60 min) was $11.5 \text{ kWh}/\text{m}^3$.

Conclusion

Electrocoagulation was assessed as a possible method for the reduction of COD, TSS, color and ammonia concentration in

paper-recycling mill wastewater. The influence of variables such as electrolysis time, voltage and initial pH on the removal of COD, TSS, color and ammonia was determined. Under optimum condition (voltage of 10 V, initial pH of 7 and operating time of 60 min), the removal efficiencies of COD, TSS, color and ammonia were 79.5%, 83.4%, 98.5 and 85.3%, respectively. In addition, other parameters including the removal efficiency of TDS and turbidity were measured at the optimum point and their values were 35% and 99%, respectively. According to the results, this method has demonstrated efficiency in the treatment of paper-recycling wastewater and can be a good alternative for other methods to be used on an industrial scale.

Acknowledgements This study was financially supported by grant No: 950704 of the Biotechnology Development Council of the Islamic Republic of Iran.

Compliance with ethical standards

Conflict of interest The authors would like to declare that there is no conflict of interest with this research and in the publication.

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