Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2015.1027960

57 (2016) 9203–9215
April

Taylor & Francis
Taylor & Francis Group

Textile wastewater treatment by application of combined chemical coagulation, electrocoagulation, and adsorption processes

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Received 17 September 2014; Accepted 6 March 2015

ABSTRACT

The aim of this work was to study the feasibility of treating real textile wastewater by combined chemical coagulation (using poly aluminum chloride (PAC) as coagulant), electrocoagulation (using aluminum electrodes), and adsorption process (using pistachio nut shell ash) to achieve the required standards. The influence of the critical operating variables such as coagulant dose, electrical potential, reaction time, and adsorbent dose on the removal efficiencies of major pollutants was determined. The results of this study were showed that removal of pollutants increased with increasing doses of PAC and applied voltage. The chemical coagulation process showed 40% of chemical oxygen demand (COD), 34% of biochemical oxygen demand (BOD_5), 23.7% TSS, and 44.5% of dye removal efficiency, even though using relatively low PAC concentration (30 mg/L). COD, BOD_5, and dye removals of 93.1, 88.8, and 98.6% were achieved by electrocoagulation process (applied voltage 60 V). In addition, COD, BOD_5, and dye removals of 98, 94.2, and 99.9% were achieved by overall combined process (chemical coagulation, electrocoagulation, and adsorption processes). The experiments demonstrated the effectiveness of combined processes for the treatment of real textile wastewaters. Consequently, combined processes are inferred to be superior to these processes alone for the removal of both organic and inorganic compounds from textile wastewater.

Keywords: Textile wastewater; Coagulation; Electrocoagulation; Adsorption; Decolorization

1. Introduction

Textile wastewater includes a large variety of dyes and chemicals additions that make the environmental challenge for textile industry not only as liquid waste but also in its chemical composition [1], hence the textile industry wastewater is one of the most polluting among all industrial sectors. Important pollutants in textile effluent are mainly recalcitrant organics, color, toxicants and inhibitory compounds, surfactants, chlorinated compounds, salts, high temperature (at least 40˚C), and high pH due to the large amount of alkali in wastewater [2]. On the other hand, textile dye wastewater is well known to contain very strong color, wide range of pH from 2 to 14, presenting large

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amount of suspended solids, chemical oxygen demand (COD) (from 50 mg/L to approximately 18,000 mg/L), high toxicity, and low biodegradable chemicals, especially the effluent from the dyeing stages of the dyeing, printing, and finishing processes [3–7]. Textile industry is among the activities that consume large capacities of fresh water on daily basis and can benefit from recycling programs.

At present time, there are various techniques available for the removal of dyes by conventional treatment technologies including biological and chemical oxidation, advanced oxidation, coagulation and adsorption processes but they cannot be effectively used individually. For example, the coagulation process effectively decolorized insoluble dyes, such as disperse dyes, but does not work well for soluble dyes. Furthermore, in coagulation process, large amount of sludge is created which may become a pollutant itself and increase the treatment cost [8,9].

Another most popular method is adsorption process. Adsorption technology for color removal is based on the affinity of various dyes for adsorbents. Activated carbon is most commonly used adsorbent and can be very effective for many dyes [10]. The limitations of this technology are the eco-friendly disposal of spent adsorbents, excessive maintenance costs, and pretreatment of wastewater to reduce the SS under acceptable range before it is fed into the adsorption column. Cost effectiveness of biological treatment process makes it attractive which can efficiently remove most of the dyes used in the textile industry because dyes generally possess high level of adsorption on to the activated sludge. However, longer duration of treatment, toxicity of dyes, large operational area, and its low biodegradability are the major limitations [11–14].

Advanced oxidation techniques result in high treatment cost and are generally used to obtain high purity grade water [8,14,15]. Generally, physicochemical treatment processes alone are not preferred, despite their effectiveness, due to the high costs involved [14]. Therefore, application of combined treatment processes can be a suitable solution for efficient removal of pollutants.

In the past, several researchers have studied the coagulation of synthetic and real textile wastewater using various inorganic chemicals (e.g. $FeCl_3$, $FeSO_4$, alum, lime, and $MgCl_2$) as well as components of biological origin, such as chitosan [16–22].

In recent years, electrochemical methods such as electrooxidation [23] and electrocoagulation have been widely used as an attractive and suitable method for the treatment of water and wastewater containing oil wastes [24,25], colloids, surfactants [26,27], dye compounds [11,28–31], and wastewaters contain heavy metals and pesticides, by virtue of various benefits including environmental compatibility, adaptability, energy efficiency, safety, selectivity, amenability to automation, and cost effectiveness [32–36].

The electrochemical treatment of wastewater is considered as one of the advanced oxidation processes, potentially a powerful method of pollution control, offering high removal efficiencies [37]. Electrocoagulation process involves the generation of coagulants *in situ* by dissolving electrically either aluminum or iron ions from aluminum or iron electrodes, respectively. In this process, the metal ions generation takes place at the anode and hydrogen gas is released from the cathode. The hydrogen gas bubbles carry the pollutant to the top of the solution where it can be more easily concentrated, collected, and removed. Various reactions take place in the electrocoagulation process, where aluminum is used as the electrode:

At the anode:

$$\mathrm{Al} \rightarrow \mathrm{Al}^{3+}_{(\mathrm{aq})} + 3\mathrm{e} \tag{1}$$

At the cathode:

$$3\mathrm{H_2O} + 3\mathrm{e} \rightarrow 3/2\mathrm{H_2} + 3\mathrm{OH}^- \tag{2}$$

The cathode may also be chemically attacked by OH^- ions generated during H_2 evolution at high pH:

$$2\mathrm{Al} + 6\mathrm{H_2O} + 2\mathrm{OH}^- \rightarrow 2\mathrm{Al(OH)}_4^- + 3\mathrm{H_2} \tag{3}$$

$Al^{3+}_{(aq)}$ and OH^- ions generated by electrode reactions (1) and (2) react to form various monomeric species such as $Al(OH)^{2+}$, $Al(OH)_2^+$, $Al_2(OH)_2^{4+}$, $Al(OH)_4^-$, and polymeric species such as $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$ and $Al_{13}(OH)_{34}^{5+}$, which transform finally into $Al(OH)_3$ according to complex precipitation kinetics [38].

Freshly formed amorphous $Al(OH)_3$ "sweep flocs" have large surface areas which are beneficial for a rapid adsorption of soluble organic compounds and trapping of colloidal particles. These flocs polymerize as:

$$n\mathrm{Al(OH)_3} \rightarrow \mathrm{Al}_n(\mathrm{OH})_{3n} \tag{4}$$

and they are easily removed from aqueous environment by sedimentation and by H_2 flotation. Secondary anodic reactions occur also during electrocoagulation process, for example, in neutral and acidic chloride solutions, native and free chlorine and hypochlorite are formed which are strong oxidants [39]. On the other hand, the aluminum hydroxide flocs normally act as adsorbents and/or traps for pollutants. Therefore, they would eliminate them from the solution [35,40].

Nevertheless, if suspended or colloidal solids were in high concentration in the wastewater, they delay the electrochemical reactions. Consequently, these materials need to be adequately removed before the electrochemical oxidation. It would be of more practical significance to the textile industry to operate the combined process in the pilot scale. Because of variability of textile wastewater composition, the traditional methods become inadequate. Various methods have been developed for treatment textile wastewater, but most of them could not be used individually in full scale because of the characteristics of textile wastewater [37,41,42].

Afterward, many combined processes are suggested in these days. One of reported combined method showed that original raw textile wastewater was treated by the combined process of chemical coagulation, electrochemical oxidation, and activated sludge [43]. Kim et al. studied pilot scale treatment of textile wastewater by combined process (fluidized biofilm process–chemical coagulation–electrochemical oxidation) [37]. In another study that was performed by Ahn et al. Fenton's oxidation and activated carbon adsorption (as the main treatment process) were employed as pretreatment processes for dyestuff wastewater having high salinity, color, and non-biodegradable organic concentrations [9]. In addition, the combined process of chemical coagulation, activated sludge filtration, and disinfection was used for treatment of textile wastewater by Nicolaou and Hadjivassilis [44].

Therefore, this study aimed to investigate the performance of conventional chemical coagulation and electrocoagulation process for pretreatment of textile wastewater prior to an adsorption process and finally to an acceptable level that can be discharged to environment with minimal adverse effects. Sequence of these processes can enhance removal efficiency of various pollutants present in textile effluents. Hence, several fundamental aspects regarding the effects of coagulant dose (poly aluminum chloride (PAC)), electrical potential, reaction time, and dose of adsorbent on the pollutants removal efficiency from real textile wastewater are explored.

2. Materials and methods

2.1. Textile wastewater

The wastewater samples were collected from the top of the settling tank in a local textile factory (with wastewater production approximately 11–13 m^3/d) situated in Iranshahr, Iran. Different classes of dyes such as acid, basic, direct, disperse, reactive and sulfur were used at various stages. The characteristics of the raw textile wastewater are presented in Table 1. Samples were collected in polypropylene bottles, shipped cold, and kept at 4°C before analysis. The effluent has been sampled at different times during this study and the initial characteristics varied with time (Table 1). This effluent initially contained high concentrations of soluble and suspended materials (278.54 ± 65.23 mg/L BOD_5, 1,346.17 ± 123.36 mg/L COD, 178.28 ± 23.82 mg/L TSS).

The continuous combined process for textile wastewater treatment consists of chemical coagulation, electrochemical oxidation, and adsorption process. A flow diagram of the continuous textile wastewater treatment system used in this study is shown in Fig. 1.

2.2. Chemical treatment (coagulation) of textile wastewater

All the chemicals used in the study were of analytical reagent (AR) grade. PAC $Al_{12}Cl_{12}(OH)_{24}$ was chosen for this study because it has been used extensively at water and wastewater treatment plants to remove solids and may function as an effective and less expensive coagulant. PAC was used in this study up to 30 mg/L (5, 10, 15, 20, 25 and 30 mg/L).

The coagulation studies were conducted in a jar test apparatus (TLT6 model) with six beakers of 2 L capacity each. Each of the beakers contained 2 L of settled textile wastewater. The coagulant (PAC) was added into the beakers without pH adjustment. Rapid mixing at 150 rpm for 4 min was followed by slow mixing at 40 rpm for 20 min. The flocs were allowed to settle for 1 h and the supernatant was carefully extracted by pipetting just below the surface of settled water. Afterward, the supernatant was filtered through a Whatman filter paper (grade 4) before examining pH, COD, biochemical oxygen demand (BOD_5), and dye concentration of the treated wastewater. Finally, after chemical coagulation, electrocoagulation process with aluminum electrodes was performed on the supernatant. All experiments were performed at room temperature (22 ± 1°C).

2.3. Electrochemical treatment (electrocoagulation) of textile wastewater

In each run, wastewater (supernatant) after chemical coagulation (first stage of treatment) was poured into the electrocoagulation cell. All experiments were performed in a bipolar batch reactor (Fig. 1), with four

Table 1
Characteristics of the raw textile wastewater used for this study

Parameters	Raw wastewater Mean ± S.D.	12 h settled wastewater Mean ± S.D.	Permissive levels (Iran standard)
Number of samples	36	36	–
Color	Blue–green	Blue–green	
Total COD (mg/L)	1,346.17 ± 123.36	1,239.3 ± 93.4	60
Total BOD (mg/L)	278.54 ± 65.23	234.38 ± 21.05	30
Total solids (mg/L)	593.55 ± 16.7	231.22 ± 15.14	–
Total suspended solids (mg/L)	178.28 ± 23.82	73.54 ± 6.4	60
Dye concentration (Pt–Co)	91.7 ± 11.4	89.6 ± 8.1	–
Conductivity (mS/cm)	1.73 ± 0.2	1.62 ± 0.19	–
pH	7.72 ± 0.03	7.64 ± 0.39	6.5–8.5

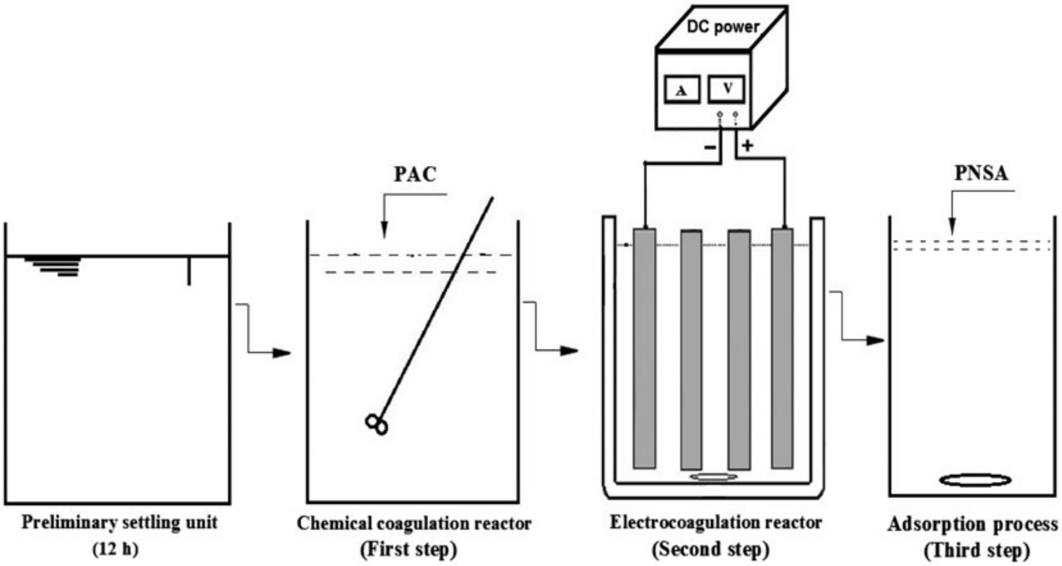

Fig. 1. The flow diagram of pilot scale combined process for textile wastewater treatment.

aluminum electrode connected in parallel. The volume (*V*) of the solution of each batch was 2 L. The active area of each electrode (plate) was 16 × 21 cm with a total area of 336 cm^2. The distance between electrodes was 1.5 cm. A power supply having an input of 220 V and variable output of 0–60 V (10, 20, 30, 40, 50, and 60 V) with maximum current of 5 A was used as direct current source. Different samples of 100 ml were taken at 15 min intervals for up to 1 h and filtered before being analyzed to determine BOD_5, COD, TS, TSS, and residual dye concentration. During the runs, the reactor unit was stirred at 120 rpm by a magnetic stirrer to allow the chemical precipitate to grow large enough for removal. Also, during electrocoagulation process, an oxide film formed at the anode and with the aim of overcoming electrode passivation the electrodes were rinsed in HCl solution (0.2 N) after each experiment and rinsed again with distilled water and finally weighted. To calculate sacrificial electrode consumption, the electrodes reweighted. These values are used in the calculations of the total operating cost. Furthermore, the electrical energy consumption per unit volume of treated wastewater has been calculated for different experimental conditions. The electrical energy consumption was calculated using the following equation [7]:

$$E = U \cdot I \cdot t/V \qquad (5)$$

where E is the energy consumption (kW h/m^3), U is the applied voltage (V), I is the current intensity (A), t

is the electrocoagulation time (h), and V is the volume of the treated wastewater (L). All analyses were conducted in duplicate for reproducibility of the experimental results.

2.4. Adsorption process

For further treatment of textile wastewater, adsorption experiments with the pistachio nut shell ash (PNSA) as a low-cost adsorbent were carried out in 1 L flasks. Each test consisted of 500 mL of supernatant (after electrocoagulation) with a desired initial concentration of adsorbent without pH adjustment and transferring it into the beaker on the magnetic stirrer. Different doses of PNSA (100–800 mg/L) was then added to the solution (according to previously study that was performed by author) [45], and the obtained suspension was immediately stirred for 60 min (shaking speed 150 rpm). After the desired contact time, the samples were withdrawn from mixture using a micropipette and centrifuged for 5 min at 5,000 rpm. After centrifuged, supernatants were analyzed for the determination of the final concentration of dye at a wavelength of 525 nm and also other mentioned parameters.

2.5. Analytical

The measurements of COD, BOD_5, TS (total solids), TSS (total suspended solids), and dye concentration followed the procedure of a standard method. COD was measured using COD reactor and direct reading spectrophotometer (DR/5000, HACH, USA). Five-day BOD_5 was determined by the manometric method with a respirometer (BSB-Controller Model 620 T (WTW)). The conductivity of solutions was measured using a Jenway Conductivity Meter (Model 4200). Turbidity of the solution was determined using a T80 UV/VIS spectrometer and pH was measured using E520 pH meter (Metrohm Herisau, Switzerland Ultra Basic, U.S.). The initial and residual dye concentrations in the solutions were analyzed using UV–Vis spectrophotometer at 525 nm wavelength (Shimadzu, Tokyo, Japan; Model 1601). The optimum wavelength was obtained by scanning the untreated samples.

3. Results and discussion

3.1. Characterization of textile wastewater

The characteristics of textile wastewater prior to any treatment, after 12 h settling time and also the guidelines from Iran for effluent discharge in the sewage urban works are presented in Table 1. BOD_5 and COD of the raw textile wastewater were 1,346.17 and 278.54 mg/L, respectively. Also, it can be noted that this effluent is not suitable for aerobic biological process due to low BOD_5/COD ratio (~0.21). Nevertheless, as it could be seen, all parameters were lowered after 12 h of preliminary settling time, however these values still very higher than recommended standard values. Consequently, the textile effluent needed to be treated before any discharge to environment.

3.2. Effect of preliminary settling

Preliminary settling process is an usual treatment method that requires no chemical addition. Although some workers realized the importance of the natural settling process, there is little information available in the literature on the effect of the preliminary settling time on pollutants removal capacity. It is clear that this wastewater with this quality is not suitable for discharge into environment.

At present study, the raw textile wastewater was allowed to settle in a preliminary settling tank before the addition of PAC as a chemical coagulant. The settling time had an effect on COD, BOD_5, TS, TSS, and dye concentration removals on the first 12 h. According to results presented in Table 1, TS and TSS were reduced from 593.55 and 178.28 mg/L to 231.22 and 73.54 mg/L (~61–66% removal efficiency), respectively. Similar results were reported by Amuda and Alade [46]. In addition, total COD was reduced in the textile wastewater from 1,346 to 1,239 mg/L (resulting in an 8% COD removal efficiency). Furthermore, concentration of BOD_5 reduced to 234 mg/L, which shows 16% reduction and also, BOD_5/COD ratio reached to ~0.18. As it mentioned earlier, low BOD_5/COD ratio indicates the necessity of an efficient physicochemical treatment for this wastewater. Similar findings were reported by Bazrafshan et al. on slaughterhouse wastewater treatment by combined chemical coagulation and electrocoagulation [47].

3.3. Effect of coagulation process (first step)

Coagulation is an essential process in the overall solid–liquid separation scheme in water and advanced wastewater treatment. Consequently, at present study chemical coagulation was employed as a pre-treatment to electrochemical oxidation. The main purpose of chemical coagulation was decreasing the pollutant loading on posttreatment. On the other hand, to obtain dischargeable effluents, PAC was intended to be used as the coagulant. Therefore, PAC was added to the wastewater to achieve particle instability and increase in the particle size, consequently achieving effective removal of organic substances present as COD and BOD_5 and other pollutants (such as TSS, turbidity, and dye). The coagulation efficiency depended on the dosage amount of coagulant. Hence for this study, the doses of PAC as coagulant were varied between 0 and 30 mg/L. During these runs, no adjustment in pH was made. Results from the jar test experiments are shown in Table 2 and Fig. 2.

It is shown that at lower doses of the PAC (5 mg/L), COD removal efficiency reached a maximum of 10.55%; BOD_5 removal efficiency reached a maximum of 7.54%; TS removal efficiency reached a maximum of 14.06% and TSS removal efficiency reached a maximum of 11.07%. As it shown in Fig. 2, the efficiency of the process increased with increasing dosages of coagulant (PAC). On the other hand, maximum removal efficiencies for all pollutants were obtained using PAC at the highest dosage of 30 mg/L.

Maximum dye removal efficiency of >44% (Fig. 2) was obtained using PAC at the dosage of 30 mg/L. The dye concentration reduction, enhance with increase in coagulant dosage. Dye concentration of effluent with coagulant dose 5 mg/L PAC was reduced from 89.6 to 80.3 mg/L (dye reduction ~10%), and by increasing the coagulant dose to 30 mg/L, the dye concentration of effluent was decreased to 49.7 mg/L (resulting in a 44.53% dye removal efficiency) that is much more than permissible level.

As seen in Fig. 2, an increase in COD, BOD_5, TS, TSS, and dye removal efficiency is noted with increasing PAC dosage, reaching nearly 40–45% for PAC dosage of 30 mg/L. Similar results were reported by Kim et al. for COD and color removal (51.2% reduction by alum as coagulant) from textile wastewater [37]. In fact, at high coagulant doses, higher metal hydroxides are produced and organic substances or soluble pollutants and colloidal particles are removed by incorporation into or sorption onto hydroxide flocs [48]. In addition, Tezcan et al. were presented alike findings regarding the application of electrocoagulation process with adding PAC as coagulant aid for treatment of vegetable oil refinery wastewater [49].

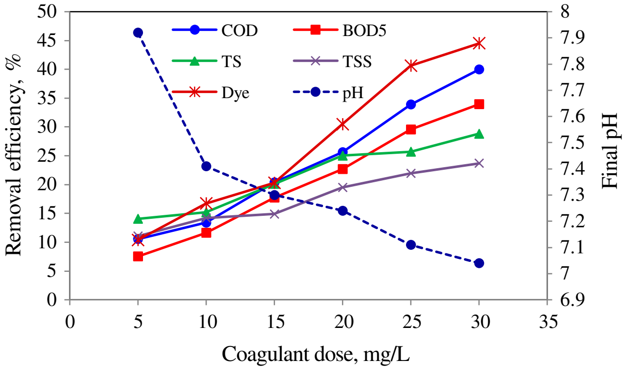

Fig. 2. Effects of coagulant dose (PAC) on pollutants removal efficiency at pilot scale coagulation process (initial pH ~ 7.6, temperature = 22 ± 1˚C).

Al-Kdasi et al. has mentioned that the chemicals and dyes that are used in textile dying process are mostly organic compounds with complex structures and hence at present study the biodegradability of the treated wastewater after chemical coagulation was determined [13]. The results revealed that the BOD_5/COD ratio was increased from 0.18 to 0.21 after the coagulation process; nevertheless, this effluent is not suitable for aerobic biological process.

In accord with the results of this study (Table 2) it can be concluded that although the removal of most pollutants from textile wastewater is high, but the concentration of pollutants in effluent of chemical coagulation process does not meet the effluent discharge standards to the environment. Thus, the effluent from conventional coagulation should be followed by another treatment process to be completed. For this

Table 2
Influence of PAC dosage on water quality parameters of coagulated mixed liquor

	Water quality parameters of treated effluent after chemical coagulation unit				
PAC dosage (mg/L)	COD (mg/L)	BOD_5 (mg/L)	TS (mg/L)	TSS (mg/L)	Dye (mg/L)
0	1,239.3	234.38	231.22	73.54	89.6
5	1,108.6	216.7	198.7	65.4	80.3
10	1,073.4	207.1	196.1	63.1	74.6
15	986.7	192.8	184.7	62.6	71.4
20	921.8	181.2	173.4	59.2	62.3
25	819.2	165.1	171.8	57.4	53.2
30	743.8	154.8	164.6	56.1	49.7

purpose, in this research, electrocoagulation was employed as a completion of treatment process to obtain discharge standards.

3.4. Effect of electrocoagulation process (second step)

Electrocoagulation has recently attracted attention as a potential technique for treating industrial effluent due to its versatility and environmental compatibility. In addition, the electrochemical treatment of wastewater is considered as one of the advanced oxidation processes, potentially a powerful method of pollution control, offering high removal efficiencies [8]. The basic premise of electrocoagulation is the production of a coagulant via an electrolysis process. By applying a current to metal plates of the required coagulant (usually aluminum or iron), metallic ions from the anode plate are released into the aqueous solution [50].

The dissolved metal ions, at an appropriate pH, can form wide ranges of coagulated species and metal hydroxides that destabilize and aggregate the suspended particles or precipitate and adsorb dissolved contaminants [51]. In addition, hydrogen gas is liberated at the cathode which causes the flotation process. The chemical reactions when using aluminum electrodes are:

$$\text{Anode}: \quad Al_{(s)} \leftrightarrow \quad Al^{3+}_{aq} + 3e^{-} \tag{6}$$

$$\text{Cathode}: \quad 3H_2O + 3e^{-} \leftrightarrow 3/2\ H_{2(g)} + 3OH^{-}_{(aq)} \tag{7}$$

$$\text{Totally}: \quad Al^{3+}_{(aq)} + 3OH^{-}_{(aq)} \leftrightarrow Al(OH)_3 \tag{8}$$

The Al^{3+} ions created can efficiently remove organic compounds, heavy metals, and even some ionic species such as fluoride and nitrate through precipitation or co-precipitation to the precipitate [52]. Then, the precipitate is captured by the $Al(OH)_3$ or polymeric aluminum hydroxide flocs and floated into a gel state.

At present work, the electrocoagulation process was employed for further treatment of effluent. In adopting the electrocoagulation process, it was intended to treat the pollutant efficiently as well as economically. Therefore, at current study the effect of critical operating parameters including applied voltage and reaction time on electrocoagulation process of textile wastewater was investigated. The results of effects of operating parameters on electrocoagulation process are shown in Table 3.

Applied voltage at the electrodes is one of the most important parameters influencing the performance and economy of the electrocoagulation process [53]. To survey the effect of applied voltage on the efficacy of electrocoagulation (after chemical coagulation), several voltages in the range of 10–60 V were applied between the electrodes in the electrocoagulation cell, and pollutants removal was determined at the conditions given in Table 3.

As presented in Table 3, the percentage of pollutants removal increases by increasing the applied voltage within the range from 10 to 60 V. These results can be explained by the fact that, increasing applied voltage or operating current would increase the dissolution rate of aluminum electrode according to Faraday's law [54] with the formation of Al^{3+} and hence the formation of $Al(OH)_3$ coagulant (reactions were presented previously). Higher rate of freshly formed amorphous $Al(OH)_3$ has large surface area on which rapid adsorption of soluble pollutants and trapping of colloidal particles take place with a consequent removal of various pollutants such as COD, BOD_5, TS, TSS, and dye from textile wastewater.

As it can be seen from Table 3, the removal efficiency of pollutants is very high and as expected, it appears that for a given time, the removal efficiency increased significantly with increase of electrical potential. As an example, COD concentration of chemical coagulation process with 5 mg/L PAC has decreased from 1,108.6 to 542.1 mg/L after electrocoagulation process with electrical potential of 10 V (approximately 51% COD removal efficiency). Again, by increasing electrical potential to 30 and 60 V for reaction time 60 min, the COD concentration in the effluent decreased to 411.8 and 152.6 mg/L. Furthermore, the COD of effluent from chemical coagulation with 30 mg/L PAC was decreased from 743.8 to 183.5 mg/L by electrocoagulation process with electrical potential of 10 V, and by increasing the electrical potential to 60 V, the COD of effluent was decreased to 51.1 mg/L (approximately 93% COD removal efficiency), that is lower than permissible level. Similar findings were reported by Phalakornkule et al. [7] on application of electrocoagulation in treating textile effluent.

Additionally according to the results of Table 3, BOD_5 of chemical coagulation process with 5 mg/L

Table 3
Influence of electrocoagulation process on effluent quality parameters (time = 60 min)

PAC dosage (mg/L)	Applied voltage (V)	Water quality parameters of treated effluent after electrocoagulation unit				
		COD (mg/L)	BOD_5 (mg/L)	TS (mg/L)	TSS (mg/L)	Dye concentration (mg/L)
5	0	1,108.6	216.7	198.7	65.4	80.3
	10	542.1	147.2	71.6	38.2	22.1
	20	478.4	130.8	66.7	36.1	18.7
	30	411.8	121.7	59.3	32.9	16.4
	40	317.4	107.3	50.2	25.7	11.6
	50	186.2	72.6	40.6	17.1	10.3
	60	152.6	43.4	29.3	16.4	9.4
10	0	1,073.4	207.1	196.1	63.1	74.6
	10	463.5	124.6	67.4	37.4	19.3
	20	371.2	118.2	62.6	33.6	18.6
	30	243.7	103.5	53.8	28.1	16.3
	40	225.6	87.3	48.3	20.4	11.8
	50	196.1	63.1	33.6	15.2	10.1
	60	123.7	40.7	22.1	13.7	6.1
15	0	986.7	192.8	184.7	62.6	71.4
	10	413.7	108.7	63.5	33.1	17.2
	20	384.3	90.4	60.4	30.6	16.7
	30	309.7	78.2	49.8	25.4	14.2
	40	194.3	62.7	44.1	19.1	10.9
	50	178.3	55.7	30.4	11.7	9.2
	60	123.8	28.9	19.7	12.9	6.0
20	0	921.8	181.2	173.4	59.2	62.3
	10	365.2	83.6	59.4	32.3	13.7
	20	296.3	76.4	57.1	27.6	12.2
	30	231.7	64.8	45.3	24.3	10.3
	40	198.6	57.1	42.7	16.2	8.2
	50	154.4	46.2	26.2	10.1	6.2
	60	102.7	23.7	16.9	9.4	3.5
25	0	819.2	165.1	171.8	57.4	53.2
	10	247.3	70.3	57.3	28.4	10.8
	20	236.9	63.6	56.2	26.1	10.1
	30	184.2	55.7	42.8	20.7	8.3
	40	151.6	46.1	38.3	14.6	5.6
	50	84.3	37.9	22.7	9.2	3.1
	60	72.6	20.3	15.1	8.1	2.4
30	0	743.8	154.8	164.6	56.1	49.7
	10	183.5	62.7	50.2	28.2	7.6
	20	168.1	52.7	48.3	23.7	6.1
	30	127.4	47.6	37.4	20.2	4.3
	40	105.3	45.4	32.9	15.1	2.4
	50	84.6	32.6	19.6	8.6	1.2
	60	51.1	17.4	9.2	6.7	0.7

PAC was reduced to 147.2 mg/L after electrocoagulation process with electrical potential of 10 V (approximately 32% BOD_5 removal efficiency), and by increasing electrical potential to 60 V, the BOD_5 concentration in the effluent decreased to 43.4 mg/L (about 80% BOD_5 removal efficiency). Furthermore, with increase in coagulant dose to 30 mg/L and increase of applied voltage to 60 V, BOD_5 concentration in effluent was reduced from 154.8 to 17.4 mg/L (on average 89% BOD_5 removal efficiency). A similar trend was seen for TS and TSS parameters. Similar findings were reported by Bazrafshan et al. [47].

Furthermore, from the results presented at Table 3, it is apparent that as the applied voltage proceeds, the dye removal efficiency increased too. For example, the dye removal efficiency at applied voltage 10 V (with coagulant dose 30 mg/L) was about 84% and by increasing the voltage to 60 V, removal efficiency was increased to more than 98%. In fact, appearance of effluent after chemical coagulation and electrocoagulation (PAC 30 mg/L and applied voltage 60 V) was very clear. In addition, the biodegradability (BOD_5/COD) of the treated wastewater after electrocoagulation process was also slightly improved, so BOD_5/COD ratio was increased from 0.21 to 0.34 (at PAC dosage 30 mg/L and applied voltage 60 V).

Electrical energy consumption is one of the most important economical parameters in the electrocoagulation process. Therefore, for the same operating conditions, after 60 min of electrocoagulation, consumption of energy and electrode material is also represented in Figs. 3 and 4. The electrical energy consumption was calculated using the related equations [7].

It can be seen from Figs. 3 and 4 that electrical energy and electrode consumption were found to increase with increasing the applied voltage as would be expected in any other electrolytic process and presented by many researches [33–36,55]. An increase in applied voltage from 10 to 60 V causes an increase in energy consumption from 0.0093 to 0.0843 kW h/L and 0.0101 to 0.0915 kW h/L of pollutants for 5 and 30 mg/L of PAC, respectively (Fig. 3). Also, an increase in applied voltage from 10 to 60 V causes an increase in electrode consumption (Fig. 4) from 0.32 to 1.38 g/L and 0.41 to 1.05 g/L of pollutants for 5 and 30 mg/L of PAC, respectively. These results are according to phenolic compounds removal efficiency by aluminum electrodes that reported by Adhoum et al. and also other researchers [40,56–60].

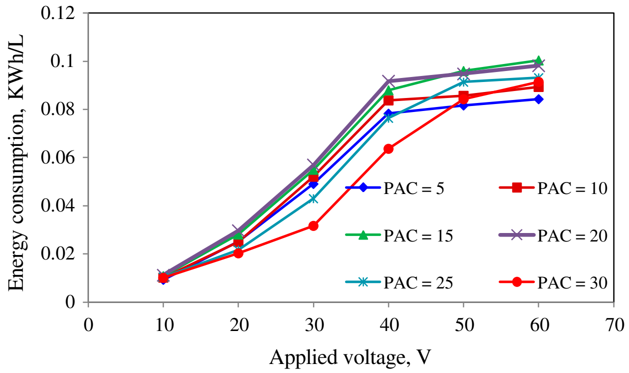

Fig. 3. Electrical energy consumption during electrocoagulation process (PAC dose = 5–30 mg/L, reaction time = 60 min).

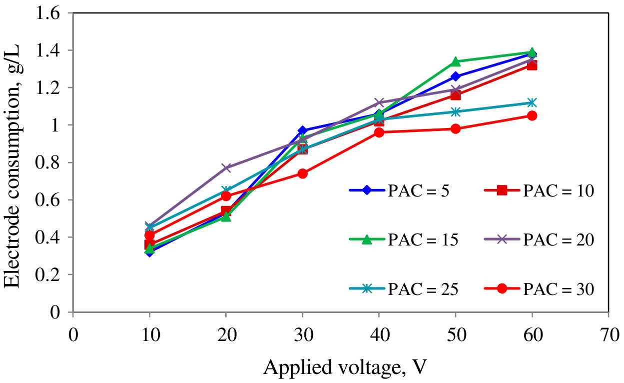

Fig. 4. Electrode consumption during electrocoagulation process (PAC dose = 5–30 mg/L, reaction time = 60 min).

3.5. Effect of adsorption process (third step)

Preliminary experiments were performed in order to investigate the effect of operating parameters and optimize the conditions of chemical coagulation and electrocoagulation process in pilot scale. The performances by the two pretreatment, namely chemical coagulation and electrocoagulation were not carried out efficiently enough for all pollutants to satisfy the Iranian guideline of effluent qualities. Additional dosage of coagulant, applied voltage, and longer reaction time were needed to keep the national guideline of the effluent qualities. Therefore, the adsorption process was employed as the final treatment step in this study.

Adsorption technology is currently being used extensively for the removal of organic and inorganic micropollutants from aqueous solutions [61]. At present study, the supernatant (after chemical coagulation with PAC 30 mg/L and electrocoagulation process at applied voltage 60 V) left after the separation of sludge was subjected to adsorption process. For the posttreatment step, the adsorbent dose was varied from 100 to 800 mg/L. COD and BOD_5 removals during the adsorption process for varying dose of activated carbon obtained from PNSA are illustrated in Fig. 5.

As it can be seen from Fig. 5, COD and BOD_5 removals are dependent on the mass of adsorbent present in the solution and it increases when the adsorbent dosage increases. On the other hand, the percentage of pollutants removal steeply increases with the adsorbent loading up to 600 mg/L. This result can be explained by the fact that the sorption sites remain unsaturated during the sorption, whereas the number of sites available for sorption site increases by increasing the adsorbent dose. The maximum

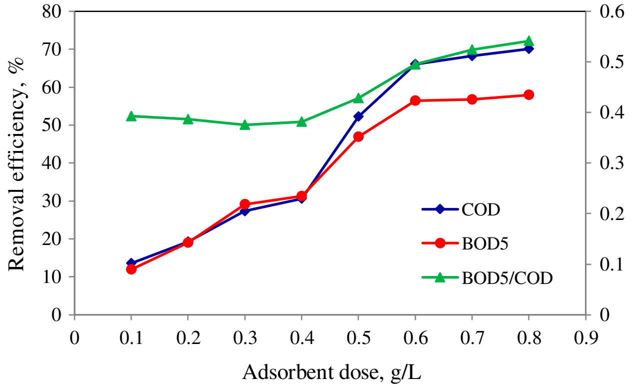

Fig. 5. Effect of adsorbent (PNSA) dose on COD and BOD_5 removal from the textile wastewater pretreated by coagulation (PAC = 30 mg/L) and electrocoagulation process (applied voltage = 60 V) at reaction time 60 min.

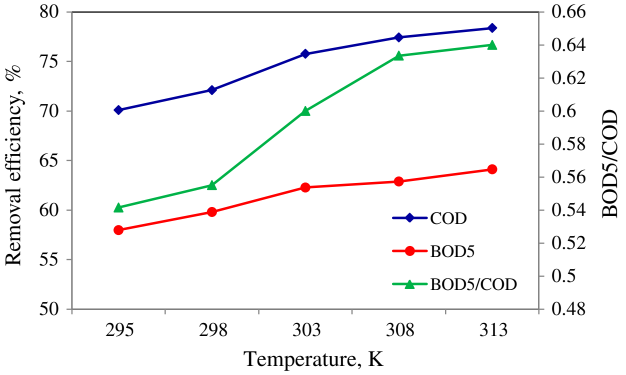

Fig. 6. Effect of temperature on COD and BOD_5 removal from the textile wastewater pretreated by coagulation (PAC = 30 mg/L) and electrocoagulation process (applied voltage = 60 V) at reaction time 60 min and adsorbent (PNSA) dosage 800 mg/L.

adsorption efficiency of COD and BOD_5 onto PNSA was found to be 70.1 and 57.98% at adsorbent concentration of 800 mg/L. Also, as it can be seen from Fig. 5, there was a no significant increase in the percentage removal of pollutants when the adsorbent concentration increases beyond the 600 mg/L. This suggests that after a certain dose of adsorbent, the maximum adsorption is attained and hence the amount of pollutants remains constant even with further addition of dose of adsorbent [62,63]. On the other hand, after a certain dose of adsorbent (600 mg/L), the maximum adsorption sets in and hence the amount of COD and BOD_5 bound to the adsorbent and the amount of COD and BOD_5 in solution remains constant even with further addition of the dose of adsorbent.

The biodegradability of the treated textile wastewater after posttreatment (adsorption process) was determined. The results revealed that the BOD_5/COD ratio was increased from 0.39 to 0.54.

According to results of this study, it can be concluded that after the combined treatment methods, the real textile wastewater can be subjected discharge to environment, safely.

Temperature has a clear effect on the adsorption capacity of the various adsorbents. To study the influence of temperature on the efficiency of adsorption process, the experiments were performed at different temperatures by maintaining the other parameters constant. For this purpose, experiments were carried out at temperatures ranging 295–313 K, pH equal 7.9, and adsorbent dosage 800 mg/L. As it can be seen from Fig. 6, the removal efficiency of COD and BOD_5 increased slightly, when the temperature was increased from 295 to 313 K.

Increasing the temperature is known to increase the rate of diffusion of the adsorbate molecules across the external boundary layer and in the internal pores of the adsorbent particle, owing to the decrease in the viscosity of the solution. Furthermore, changing temperature will change the equilibrium capacity of the adsorbent for a particular adsorbate. Patel and Vashi [10] investigated the effect of temperature (ranging 300–360 K) on removal of COD, BOD_5, and color from textile wastewater. Their findings showed that the removal efficiency continuously increases with increase in temperature [10]. Furthermore, similar findings were reported by other researchers [63,64].

4. Conclusions

In this study, chemical coagulation using PAC, electrocoagulation process with aluminum electrodes, and adsorption process using PNSA of textile wastewater was investigated. The effects of the different operational parameters on the removal of pollutants were analyzed. The following conclusions can be reached from the results obtained in this work:

(1) Preliminary settling time was investigated and found to be important operational parameter for effective treatment of real textile wastewater.
(2) A preliminary settling time of 12 h had an effect on the COD, BOD_5, TS, TSS, and dye concentration with removal efficiency up to 7.94, 15.85, 61.04, 58.75, and 2.29%, respectively.
(3) According to the findings obtained from this study, the removal efficiencies increased by

increasing the coagulant dose, applied electrical potential, and adsorbent dose. Moreover, the energy consumption increased by increasing the applied electrical potential.

(4) Finally, it can be concluded that the combined process chemical coagulation, electrocoagulation, and adsorption process has the potential to be utilized for the cost-effective removal of pollutants from real textile wastewater.

Acknowledgments

The authors are grateful for the financial support provided by the health research deputy of Zahedan University of Medical Sciences (Project No. 5670).

References

[1] M.C. Venceslau, S. Tom, J.J. Simon, Characterization of textile wastewaters—A review, Environ. Technol. 15 (1994) 917–929.

[2] S. Şen, G.N. Demirer, Anaerobic treatment of real textile wastewater with a fluidized bed reactor, Water Res. 37 (2003) 1868–1878.

[3] F. El-Gohary, A. Tawfik, Decolorization and COD reduction of disperse and reactive dyes wastewater using chemical-coagulation followed by sequential batch reactor (SBR) process, Desalination 249 (2009) 1159–1164.

[4] W.J. Lau, A.F. Ismail, Polymeric nanofiltration membranes for textile dye wastewater treatment: Preparation, performance evaluation, transport modelling, and fouling control—A review, Desalination 245 (2009) 321–348.

[5] I. Ciabatti, F. Tognotti, L. Lombardi, Treatment and reuse of dyeing effluents by potassium ferrate, Desalination 250 (2010) 222–228.

[6] E. Debik, G. Kaykioglu, A. Coban, I. Koyuncu, Reuse of anaerobically and aerobically pre-treated textile wastewater by UF and NF membranes, Desalination 256 (2010) 174–180.

[7] C. Phalakornkule, S. Polgumhang, W. Tongdaung, B. Karakat, T. Nuyut, Electrocoagulation of blue reactive, red disperse and mixed dyes, and application in treating textile effluent, J. Environ. Manage. 91 (2010) 918–926.

[8] V. Khandegar, A.K. Saroha, Electrocoagulation for the treatment of textile industry effluent—A review, J. Environ. Manage. 128 (2013) 949–963.

[9] D.H. Ahn, W.S. Chang, T.I. Yoon, Dyestuff wastewater treatment using chemical oxidation, physical adsorption and fixed bed biofilm process, Process Biochem. 34(5) (1999) 429–439.

[10] H. Patel, R.T. Vashi, Treatment of textile wastewater by adsorption and coagulation, E-J. Chem. 7(4) (2014) 1468–1476.

[11] S.I. Abo-Elela, F.A. El-Gohary, H.L. Ali, R.S. Abdel-Wahaab, Treatability studies of textile wastewater, Environ. Technol. Lett. 9 (1998) 101–109.

[12] S.H. Lin, M.L. Chen, Treatment of textile wastewater by chemical methods for reuse, Water Res. 31 (1997) 868–876.

[13] A. Al-kdasi, A. Idris, K. Saed, C.T. Guan, Treatment of textile wastewater by advanced oxidation processes—A review, Global Nest: Int. J. 6(3) (2004) 222–230.

[14] A.K. Verma, R.R. Dash, P. Bhunia, A review on chemical coagulation/flocculation technologies for removal of colour from textile wastewaters, J. Environ. Manage. 93 (2012) 154–168.

[15] O.T. Can, M. Bayramoglu, M. Kobya, Decolorization of reactive dye solutions by electrocoagulation using aluminum electrodes, Ind. Eng. Chem. Res. 42 (2003) 3391–3396.

[16] M.A.A. Hassan, T.P. Li, Z.Z. Noor, Coagulation and flocculation treatment of wastewater in textile industry using chitosan, J. Chem. Nat. Resour. Eng. 4 (2009) 43–53.

[17] J. Mo, J.E. Hwang, J. Jegal, J. Kim, Pretreatment of a dyeing wastewater using chemical coagulants, Dyes Pigm. 72 (2007) 240–245.

[18] H. Selcuk, Decolorization and detoxification of textile wastewater by ozonation and coagulation processes, Dyes Pigm. 64 (2005) 217–222.

[19] N. Azbar, T. Yonar, K. Kestioglu, Comparison of various advanced oxidation processes and chemical treatment methods for COD and color removal from a polyester and acetate fiber dyeing effluent, Chemosphere 55 (2004) 35–43.

[20] A. Szygula, E. Guibal, M. Palacin, N. Adhoum, L. Monser, N. Bellakhal, J.E. Belgaied, Treatment of electroplating wastewater containing Cu^{2+}, Zn^{2+} and Cr(VI) by electrocoagulation, J. Hazard. Mater. B112 (2004) 207–213.

[21] D. Georgiou, A. Aivazidis, J. Hatiras, K. Gimouhopoulos, Treatment of cotton textile wastewater using lime and ferrous sulfate, Water Res. 37 (2003) 2248–2250.

[22] B.H. Tan, T.T. Teng, A.K.M. Omar, Removal of dyes and industrial dye wastes by magnesium chloride, Water Res. 34 (2000) 597–601.

[23] U. Tezcan, U. Altay, A.S. Koparal, U. Bakir, Ogutveren, Complete treatment of olive mill wastewaters by electrooxidation, Chem. Eng. J. 139 (2008) 445–452.

[24] N. Biswas, G. Lazarescu, Removal of oil from emulsions using electrocoagulation, Int. J. Environ. Stud. 38 (1991) 65–75.

[25] V.E. Cenkin, A.N. Belevstev, Electrochemical treatment of industrial wastewater, Eff. Water Treat. J. 25 (7) (1985) 243–249.

[26] P. Cañizares, C. Jiménez, F. Martínez, M.A. Rodrigo, C. Sáez, The pH as a key parameter in the choice between coagulation and electrocoagulation for the treatment of wastewaters, J. Hazard. Mater. 163 (2009) 158–164.

[27] S. Khoufi, F. Feki, S. Sayadi, Detoxification of olive mill wastewater by electrocoagulation and sedimentation processes, J. Hazard. Mater. 142 (2007) 58–67.

[28] M.S. Mahmoud, J.Y. Farah, T.E. Farrag, Enhanced removal of methylene blue by electrocoagulation using iron electrodes, Egypt. J. Pet. 22 (2013) 211–216.

[29] U. Tezcan Un, E. Aytac, Electrocoagulation in a packed bed reactor-complete treatment of color and

cod from real textile wastewater, J. Environ. Manage. 123 (2013) 113–119.

[30] N. Daneshvar, H. Ashassi-Sorkhabi, M.B. Kasiri, Decolorization of dye solution containing Acid Red 14 by electrocoagulation with a comparative investigation of different electrode connections, J. Hazard. Mater. 112 (2004) 55–62.

[31] M. Kobya, O.T. Can, M. Bayramoglu, M. Sozbir, Operating cost analysis of electrocoagulation of textile dye wastewater, Sep. Purif. Technol. 37 (2004) 117–125.

[32] E. Bazrafshan, A.H. Mahvi, M.A. Zazouli, Removal of zinc and copper from aqueous solutions by electrocoagulation technology using iron electrodes, Asian J. Chem. 23(12) (2011) 5506–5510.

[33] E. Bazrafshan, A.H. Mahvi, S. Nasseri, A.R. Mesdaghinia, Performance evaluation of electrocoagulation process for removal of chromium (VI) from synthetic chromium solutions using iron and aluminum electrodes, Turkish. J. Eng. Environ. Sci. 32(2) (2008) 59–66.

[34] E. Bazrafshan, A.H. Mahvi, S. Nasseri, M. Shaighi, Performance evaluation of electrocoagulation process for Diazinon removal from aqueous environment by using iron electrodes, Iran. J. Environ. Health Sci. Eng. 4 (2007) 127–132.

[35] E. Bazrafshan, A.H. Mahvi, S. Nasseri, A.R. Mesdaghinia, F. Vaezi, Sh. Nazmara, Removal of cadmium from industrial effluents by electrocoagulation process using iron electrodes, Iran. J. Environ. Health Sci. Eng. 3 (2006) 261–266.

[36] J. Nouri, A.H. Mahvi, E. Bazrafshan, Application of electrocoagulation process in removal of zinc and copper from aqueous solutions by aluminum electrodes, Int. J. Environ. Res. 4(2) (2010) 201–208.

[37] T.H. Kim, C. Park, J. Lee, E.-B. Shin, Pilot scale treatment of textile wastewater by combined process (fluidized biofilm process-chemical coagulation–electrochemical oxidation), Water Res. 36 (2002) 3979–3988.

[38] A. Gürses, M. Yalçin, C. Doğar, Electrocoagulation of some reactive dyes: A statistical investigation of some electrochemical variables, Waste Manage. 22 (2002) 491–499.

[39] A.G. Vlyssides, D. Papaioannou, M. Loizidoy, P.K. Karlis, A.A. Zorpas, Testing an electrochemical method for treatment of textile dye wastewater, Waste Manage. 20 (2000) 569–574.

[40] N. Adhoum, L. Monser, N. Bellakhal, J.E. Belgaied, Treatment of electroplating wastewater containing Cu^{2+}, Zn^{2+} and Cr(VI) by electrocoagulation, J. Hazard. Mater. 112 (2004) 207–213.

[41] P.M. Sutton, P.N. Mishra, Activated carbon based biological fluidized beds for contaminated water and wastewater treatment: A state-of-the-art review, Water Sci. Technol. 29 (1994) 309–317.

[42] T. Robinson, G. McMullan, R. Marchant, P. Nigam, Remediation of dyes in textile effluent: A critical review on current treatment technologies with a proposed alternative, Bioresour. Technol. 77 (2001) 247–255.

[43] S.H. Lin, C.F. Peng, Continuous treatment of textile wastewater by combined coagulation, electrochemical oxidation and activated sludge, Water Res. 30 (1996) 587–592.

[44] M. Nicolaou, I. Hadjivassilis, Treatment of wastewater from the textile industry, Water Sci. Technol. 25(1) (1992) 31–35.

[45] E. Bazrafshan, F. Kord Mostafapour, A.H. Mahvi, Phenol removal from aqueous solutions using pistachio-nut shell ash as a low cost adsorbent, Fresen. Environ. Bull. 21(10) (2012) 2962–2968.

[46] O.S. Amuda, A. Alade, Coagulation/flocculation process in the treatment of abattoir wastewater, Desalination 196 (2006) 22–31.

[47] E. Bazrafshan, F. Kord Mostafapour, M. Farzadkia, K.A. Ownagh, A.H. Mahvi, Slaughterhouse wastewater treatment by combined chemical coagulation and electrocoagulation process, Plos One 7(6) (2012) 1–8.

[48] S. Demirci, B. Erdogan, R. Ozcimder, Wastewater treatment at the petroleum refinery, Kirikkale, Turkey using some coagulants and Turkish clays as coagulant aids, Water Res. 32(11) (1998) 3495–1499.

[49] U. Tezcan Un, A.S. Koparal, U. Bakir Ogutveren, Electrocoagulation of vegetable oil refinery wastewater using aluminum electrodes, J. Environ. Manage. 90 (2009) 428–433.

[50] T. Mariam, L.D. Nghiem, Landfill leachate treatment using hybrid coagulation–nanofiltration processes, Desalination 250 (2010) 677–681.

[51] P. Cañizares, M. Carmona, J. Lobato, F. Martínez, M.A. Rodrigo, Electrodissolution of aluminum electrodes in electrocoagulation processes, Ind. Eng. Chem. Res. 44 (2005) 4178–4185.

[52] M.M. Emamjomeh, M. Sivakumar, Review of pollutants removed by electrocoagulation and electrocoagulation/flotation processes, J. Environ. Manage. 90(5) (2009) 1663–1679.

[53] M. Mollah, R. Schennach, J.R. Parga, D.L. Cocke, Electrocoagulation (EC)—Science and applications, J. Hazard. Mater. 84 (2001) 29–41.

[54] P.K. Holt, G.W. Barton, C.A. Mitchell, The future for electrocoagulation as a localised water treatment technology, Chemosphere 59 (2005) 355–367.

[55] C.A. Martínez-Huitle, E. Brillas, Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods: A general review, Appl. Catal. B: Environ. 87 (2009) 105–145.

[56] E. Bazrafshan, F. Kord Mostafapour, M.M. Soori, A.H. Mahvi, Application of combined chemical coagulation and electrocoagulation process to carwash wastewater treatment, Fresen. Environ. Bull. 21(9a) (2012) 2694–2701.

[57] E. Bazrafshan, H. Biglari, A.H. Mahvi, Phenol removal by electrocoagulation process from aqueous solutions, Fresen. Environ. Bull. 21(2) (2012) 364–371.

[58] E. Bazrafshan, H. Biglari, A.H. Mahvi, Humic acid removal from aqueous environments by electrocoagulation process using iron electrodes, E-J. Chem. 9(4) (2012) 2453–2461.

[59] E. Bazrafshan, K.A. Ownagh, A.H. Mahvi, Application of electrocoagulation process using iron and aluminum electrodes for fluoride removal from aqueous environment, E-J. Chem. 9(4) (2012) 2297–2308.

[60] E. Bazrafshan, H. Moein, F. Kord Mostafapour, Sh. Nakhaie, Application of electrocoagulation process for dairy wastewater treatment, J. Chem. 2013 (2013) 1–8.

[61] M. Ahmaruzzaman, Adsorption of phenolic compounds on low-cost adsorbents: A review, Adv. Colloid Interface Sci. 143 (2008) 48–67.

[62] P. Chakravarty, N.S. Sarma, H.P. Sarma, Removal of lead(II) from aqueous solution using heartwood of *Areca catechu* powder, Desalination 256 (2010) 16–21.

[63] E. Bazrafshan, A.A. Zarei, H. Nadi, M.A. Zazouli, Adsorptive removal of methyl orange and reactive red 198 by *Moringa Peregrina* ash, Indian J. Chem. Technol. 21 (2014) 105–113.

[64] E. Bazrafshan, F. Kord Mostafapour, S. Rahdar, A.H. Mahvi, Equilibrium and thermodynamics studies for decolorization of Reactive Black 5 (RB5) by adsorption onto MWCNTs, Desalin. Water Treat. 2014 (in press).