Membrane Water Treatment, *Vol. 9, No. 1 (2018) 9-15*
DOI: https://doi.org/10.12989/mwt.2018.9.1.009

Study of chemical coagulation conditions for a disperse red dye removal from aqueous solutions

Mohammed Tiaiba[1], Belkacem Merzouk[*2,3], Mohammed Mazour[1], Jean P. Leclerc[4] and François Lapicque[4]

[1]Laboratoire d'Hydrologie Appliquée et Environnement, Centre Universitaire d'Ain Témouchent, Ain Témouchent, Algérie
[2]Département Hydraulique, Faculté de Technologie, Université Mohamed Boudiaf de M'sila, B.P. 166, Ichbilia, M'sila, 28000 Algérie
[3]Laboratoire 3BS, Université de Bejaia, Bejaia 06000, Algérie
[4]Laboratoire Réactions et Génie des Procédés (LRGP) UMR 7274, CNRS, Université de Lorraine, 1 rue Grandville, BP 20451, 54001 Nancy Cedex, France

(Received June 23, 2016, Revised January 26, 2017, Accepted May 25, 2017)

Abstract. Coagulation process using aluminum sulfate ($Al_2(SO_4)_3$) and ferric chloride ($FeCl_3$) was employed as a treatment method for decolorization of a synthetic textile wastewater containing red dye in this paper. Factors such as initial pH, coagulant dosage, initial concentration, conductivity and mixing conditions that influence color removal efficiency were experimentally tested. It was found that $Al_2(SO_4)_3$ is more efficient than $FeCl_3$ as coagulant. When 40 mgL^{-1} aluminum sulfate was used, results showed that color induced by the red dye was efficiently removed (> 90 %) and was obtained in a large range of initial pH from 4 to 8 with, and for a dye concentration lower than 235 mg L^{-1}. After addition of the coagulant, the medium had to be mixed for 30 min at 60 rpm, then allowed to settle for 40 min. The effects of water conductivity in the range 0.035 - 2.42 mS cm^{-1} and dye concentration up to 380 mg L^{-1} were also followed and discussed.

Keywords: coagulation; red dye; aluminum sulfate; ferric chloride; decolorization

1. Introduction

The textile industry is known as a water intensive sector which employs a wide variety of processes. The dyeing step in the textile production has the largest risk for the environment due to high concentrations of organic dyes, additives and salts used (Van der Bruggen *et al.* 2004, Chafi *et al.* 2011). Most of the time, this process represents the major part of the water consumption and generates wastewaters characterized by high chemical oxygen demand (COD), high dissolved and suspended solids, and high color contents (Pela and Tokat 2002, Barka *et al.* 2011).

The dyes are one important part of the pollution problem as it is estimated that 50% of their amount is not fixed on fibers and remain free in the wastewater generated. A number of treatment procedures have been proposed to solve this problem, e.g., adsorption, electrocoagulation, chemical coagulation, electrochemical oxidation, microfiltration, nanofiltration filtration and biological treatment (Rossini *et al.* 1999, Kim *et al.* 2003, Verma *et al.* 2012, Şengil and Özdemir 2012, Verma *et al.* 2014, Zerrouki *et al.* 2014, Tahri *et al.* 2012). First, activated carbon adsorption presents additional cost and difficulty in the regeneration process, coupled with a high waste disposal cost. Similarly, advanced oxidation processes such as ozonization, UV and ozone/UV combined oxidation, photo-catalysis (UV/TiO_2), Fenton reactive and ultrasonic oxidation are not yet economically viable. Also, biological methods cannot be applied to most textile wastewaters due to the toxicity of most commercial dyes to the microorganisms used in the process (Shen and Wang 2002, Rasorio *et al.* 2002, Stanislaw *et al.* 2001, Arslan *et al.* 2002, Can *et al.* 2006, Joseph *et al.* 2012). Chemical coagulation (CC) may be induced by the destabilization of the electrostatic interactions between the dye molecules and water, upon addition of a chemical reagent, namely, the coagulant. Many chemical coagulants are available for wastewater treatment, such as aluminum chloride, alum and derivatives, lime or ferric chloride, magnesium chloride ($MgCl_2$), polyaluminium chloride (PACl) (Tan *et al.* 2000, Matilainen *et al.* 2010, Moghaddam *et al.* 2010, Verma *et al.* 2014). However, $Al_2(SO_4)_3$ and, to a lesser extent, $FeCl_3$ are among the most popular. The coagulation step is followed by flocculation, which can be favored by additives, such as synthetic polymers with a linear structure and a high molecular weight. Dye removal by coagulation is not based on the partial decomposition of dye compounds, thus no potentially harmful and toxic intermediates are produced. Furthermore, this process can be used in large-scale operation with relatively high operability and cost effectiveness (Kim *et al.* 2004, Papi´c *et al.* 2004, Selçuk *et al.* 2004). The majority of the studies focus itself on the study of the various types of coagulants and flocculants. Few studies have focused on the effects of operating parameters such as the mixing rate, mixing time and settling time (Rossini *et al.* 1999, Lee *et al.* 2006, Guida *et al.* 2007, Joo *et al.* 2007, Mo *et al.* 2007). On the basis of the above discussion, the main objective of this study was to examine the chemical coagulation performance of aluminum sulfate

---

∗Corresponding author, Professor
E-mail: mbelka01@yahoo.fr

ISSN: 2005-8624 (Print), 2092-7037 (Online)

and ferric chloride at various dosages to removal color and to reduce COD under different operating conditions, such as pH conditions, initial concentrations, mixing conditions, conductivity and settling time: the optimal parameters for COD reduction and color removal for a synthetic and real textile wastewater could then be determined.

2. Materials and methods

2.1 Coagulation experiments

Coagulation process was investigated in the batch mode by running series of six experiments in parallel in a multiple stirrer jar-test apparatus. Two coagulants were compared: aluminum sulfate ($Al_2(SO_4)_3$, $18H_2O$) and ferric chloride ($FeCl_3$, $6H_2O$). The experiments were carried out at room temperature (24±2°C), using a 1000 mL wastewater sample, by applying six different coagulant concentrations ranging from 20 mg L^{-1} to 120 mg L^{-1}. During the tests, coagulant has been dispersed rapidly in a first time with rapid agitation of 180 rpm for 3 min; these stirring conditions were kept constant for all the experiments. According to the literature, this phase would have little influence on pollution abatement (Semmens and Field 1980, Lefebvre and Legube 1990). As a matter of fact, duration of the stirring phase can vary from a few seconds to 10 minutes, but the low effect of the time period is explained by the fact that the hydrolysis reactions of the coagulant are usually very fast processes. Then, coagulation was studied at slower stirring speed: 30 rpm, 40 rpm and 60 rpm, respectively, for two mixing time periods at 20 min and 30 min. Once stirring was stopped, settling could occur. The settling time was varied between 10 min and 60 min. After settling, 20 mL of supernatant fluid was taken out at a fixed distance of 2 cm below the air-liquid interface with a syringe for pH, color and COD measurements.

2.2 Chemicals and methods

Experiments were carried out using a red dye solution (Fig. 1) with a total concentration (C_i) that was varied between 25 mg L^{-1} and 380 mg L^{-1}. This dye belongs to the dispersive dye class (Kim *et al.* 2002, Chafi *et al.* 2011), which covers nearly insoluble organic and non-ionic compounds that are however applied in aqueous solution using the simple immersion technique (Kim *et al.* 2002). The dye used and its concentration are typical of wastewater from Algerian textile factories. Synthetic solutions were prepared by mixing the dye in tap water in a wastewater tank. Solution conductivity (κ) and pH were measured using a CD810 conductimeter (Radiometer Analytical, France) and a ProfilLine pH197i pHmeter (WTW, Germany). Dye concentration was estimated from its absorbance characteristics in the UV-vis range (200-800 nm), at the wavelength corresponding to the maximum intensity (λ_{max}=450 nm) using a UV-vis spectrophotometer (Pye Unicam, SP 8-400, UK). Measurements were carried out in triplicate. COD was measured using the standard closed reflux colorimetric method. The COD of the textile wastewater used was near 2500 mg L^{-1}. The pH of wastewater before coagulation was varied from 2.65 to 9.1 using aliquots of 0.1 M H_2SO_4 or NaOH solutions (Jiang and Graham 1996, Kim *et al.* 2002, Kobya *et al.* 2003, Alinsafi *et al.* 2005, Zodi *et al.* 2011).

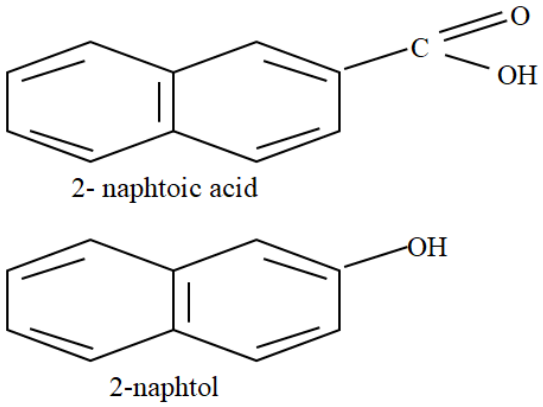

Fig. 1 Molecular structure of the constituents of the red dye

Samples were collected and filtered using 0.45 µm membrane filters for residual dye measurements. Absorbance at 450 nm was measured every 10 min in order to follow the evolution of color removal vs. time. For COD, only the initial value before coagulation and the final value after settling were measured. COD and color removal efficiencies (Y_{COD}, Y_{COL}) were therefore expressed as a percentage and defined as

$$Y_{COL}\,(\%) = \frac{C_i - C_f}{C_i}.100 \tag{1}$$

$$Y_{COD} = \frac{COD_i - COD_f}{COD_i}.100 \tag{2}$$

In Eq. (1), C_i and C_f are the initial and final dye concentrations expressed in mg L^{-1} of the jar test, respectively. The same subscripts, "i" and "f", are used to distinguish initial and final COD values in Eq. (2). Statistical analysis on replicated measurements gave access to Y_{COL} values with ± 0.2% and Y_{COD} values ± 1% accuracy.

3. Results and discussions

3.1 Effects of mixing rate, mixing time and settling time on the color removal efficiency

In order to analyze the respective effects of mixing rate and mixing time on the color removal efficiency, Fig. 2 illustrates the influence of mixing rate in the range 30-60 rpm as a function of settling time for 20 and 30 min mixing time, respectively. Fig. 2 presents the particular case of 20 mg L^{-1} $Al_2(SO_4)_3$ as a coagulant with a 100 mg L^{-1} initial dye concentration at pH 6.5 and κ=2.4 mS cm^{-1}: this example is representative of the trends observed for other dye and coagulant concentrations.

Experimental results show that the color removal efficiency Y_{COL} increased with mixing time and settling time, but also with the mixing rate in the moderate range studied, as only moderate rotation speeds were studied. The

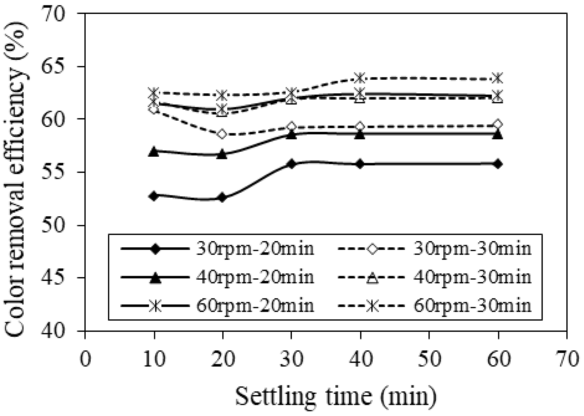

Fig. 2 Effects of mixing speed, mixing time and settling time on the color removal efficiency: C_i=100 mg/L, initial pH 6.45, κ=2.4 mS/cm (aluminum sulfate)

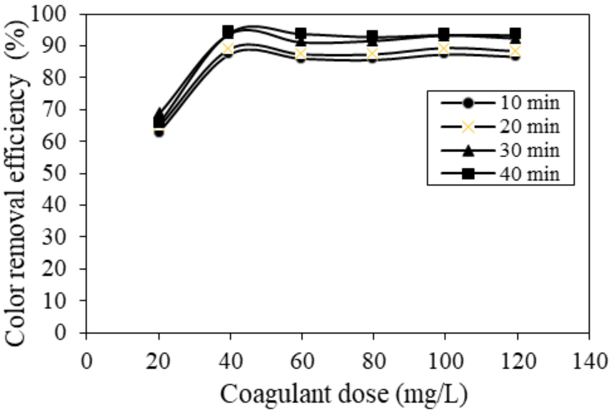

Fig. 3 Effect of coagulant dosage and settling time on the color removal efficiency: C_i=100 mg/L, initial pH 6.5, κ=2.4 mS/cm, mixing speed=60 rpm, mixing time=30 min (aluminum sulfate)

efficiency enhancement by increased rotation rate over 40 rpm became however insignificant with a 30 min-long mixing period. Similarly, mixing at 60 rpm allowed Y_{COL} to be improved by only 2%. Settling time is the parameter that exhibits the lowest effect over 10 min: only a slight increase was observed for settling time at 40 min or below. In Fig. 2, it must be mentioned that the maximum color removal efficiency was 63.8% upon 30 min mixing at 60 rpm and 40 min settling. This efficiency constitutes an insufficient result that must still be improved, for example by changing the coagulant type and its dosage.

3.2 Effects of coagulant type and dosing on the color removal efficiency

3.2.1 Aluminum sulfate

The effect of aluminum sulfate dosage is shown in Fig. 3. All parameters, except the coagulant dosage, were kept constant. It can be clearly seen that color removal increased with higher aluminum contents until a plateau value could be attained. As a matter of fact, for an initial pH value of 6.5, the color removal rose steeply from 20 to 40 mg L^{-1} coagulant added, whereas further addition of aluminum sulfate resulted only in insignificant improvement of performance, giving a total color removal near 94% after 40 min settling with a stirring speed of 60 rpm. It is worth of note that the settling time had nearly no influence on color removal, as shown in Fig. 2, whatever the coagulant dose. In Fig. 3, the color removal seems to exhibit a flat maximum for coagulant dose ranging from 20 to 60 mg L^{-1}, especially upon 30 minute-long settling. The highest efficiency at pH 6.5 is therefore obtained with 40 mg L^{-1} aluminum sulfate. The existence of a flat maximum is not surprising, since it is actually known that too large coagulant dosages can be detrimental because of extensive precipitation of aluminum hydroxide in excess, which slightly hinders the coagulation (Duan and Gregory 2003, Verma *et al.* 2014).

3.2.2 Ferric chloride

The study of the effect of the iron chloride dosage on color removal has been undertaken using a method similar to that used for $Al_2(SO_4)_3$ in the previous section. First, the effect of settling time has been shown to be negligible in the range studied, as for aluminum sulfate. However, the effect of coagulant dose strongly differs from the former case, as seen in Fig. 4. Indeed, it is obvious that the amount of ferric chloride required for achieving an Y_{COL} value of 90% is far higher than with aluminum sulfate (Fig. 3): 120 mg/L iron chloride are needed in comparison with only 40 mg L^{-1} aluminum salt. The above mass concentrations correspond to very different metal form concentrations, at 0.44 mM for iron chloride, and only 0.06 mM for trivalent Al. From Fig. 4, it is clear that coagulation by charge neutralization prevails up to 60 mg L^{-1}, which corresponds to a maximum color removal efficiency of about 74%. However, the weak maximum due to charge reversal and particle destabilization is replaced for $FeCl_3$ by a short plateau region between 60 and 80 mg L^{-1} coagulant, followed by an increase in Y_{COL} that can be attributed to a hydroxide precipitation-driven coagulation mechanism (Duan and Gregory 2003).

The data obtained suggest that color removal does not correspond a simple complexation of the dye molecule with the metal species with a given stoichiometry since Fe and Al species act in very different manner, but obeys to more complex phenomena, involving likely the morphology of the hydroxides formed: *Al* and *Fe* hydroxides have actually very different morphology and texture. Nevertheless, due to the poor performance of ferric chloride as a coagulant, only aluminum sulfate has been used in the following sections.

3.3 Effect of inlet solution pH on the color removal efficiency

The inlet solution pH of wastewater is a key parameter in the coagulation process (Olthof and Eckenfelder 1976, Szygu1a *et al.* 2009, Li *et al.* 2010, Matilainen *et al.* 2010). The use of coagulant at its optimum pH allows maximum pollutant removal (Amokrane *et al.* 1997, Szygu1a *et al.* 2009, Moghaddam *et al.* 2010, Joseph *et al.* 2012). Fig. 5 gives the solubility diagram for aluminum hydroxide, $Al(OH)_{3(s)}$, assuming only mononuclear species. $Al(OH)_3$ formed at pH 5.0-8.5 interval. The solubility boundary denotes the thermodynamic equilibrium that exists between the dominant aluminum species at a given pH and solid aluminum hydroxide. The minimum solubility occurs at

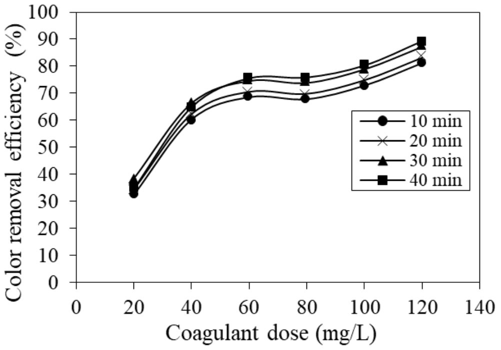

Fig. 4 Effect of coagulant dose and settling time on the color removal efficiency: C_i=100 mg/L, initial pH 6.5, κ=2.4 mS/cm, mixing speed=60 rpm, mixing time=30 min (ferric chloride)

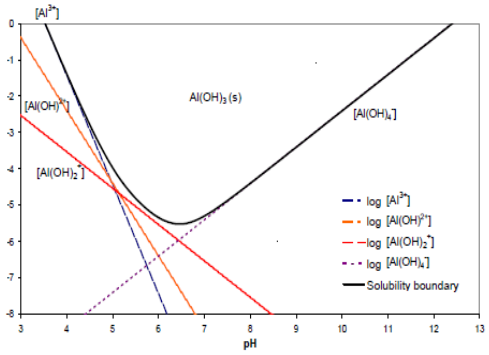

Fig. 5 Solubility diagram of aluminum hydroxide $Al(OH)_{3(s)}$ considering only mononuclear aluminum species (Holt *et al.* 2002)

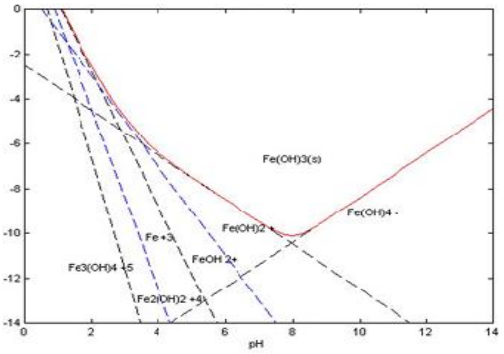

Fig. 6 Speciation diagram for iron (Cook *et al.* 2012)

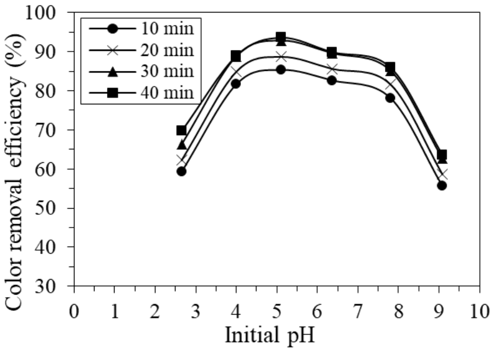

Fig. 7 Effect of initial pH on the color removal efficiency: C_i=100 mg/L, mixing speed=60 rpm, mixing time=30 min, 40 mg/L of $Al_2(SO_4)_3$, κ=2.4 mS/cm

Table 1 Evolution of pH values during coagulation process during 40 min settling time for different values of initial pH: C_i=100 mg/L, mixing speed=60 rpm, mixing time=30 min, 40 mg/L of $Al_2(SO_4)_3$, κ=2.4 mS/cm

Initial pH	2.7	4.0	5.1	6.4	7.8	9.1
Final pH	3.0	4.2	4.5	5.7	6.0	6.8

approximately pH 6.5, with solubility increasing as the solution becomes more acidic or alkaline. However, as the aluminum concentration increases, polynuclear aluminum complexes are formed and aluminum hydroxide precipitates. Examination of activity–pH diagram for Al(III) species in equilibrium with $Al(OH)_{3(s)}$ shows that dominant Al(III) species is in the form of $Al(OH)_{3(s)}$ in the 5.0-8.5 pH range.

Correspondingly for iron (Fig. 6), after the electrogeneration of ferric ions, monomeric ions, ferric hydroxocomplexes with OH^- ions, and polymeric species may be formed. These species include $Fe(OH)_2^+$, $Fe_2(OH)_2^{4+}$, $Fe(OH)_4^-$, $Fe(H_2O)_5OH_2^+$, $Fe(H_2O)_4(OH)_2^+$, $Fe(H_2O)_8(OH)_2^{4+}$, and $Fe_2(H_2O)_6(OH)_4^{2+}$. As with aluminum, these species will further react to form $Fe(OH)_3$ (Kobya *et al.* 2003). One of the main factors effecting the formation of the metal complexes is solution pH. The dominant species in solutions with a pH value above 9 are $Al(OH)_4^-$ and $Fe(OH)_4^-$ (Figs. 5 and 6). These forms don’t contribute to floc formation (Chafi *et al.* 2011, Holt *et al.* 2002, Adjeroud *et al.* 2015).

To determine the optimum pH of the coagulation process, 40 mg aluminum sulfate, on the basis of Fig. 3, have been added to each jar containing 1000 mL of wastewater at different pH values, adjusted using H_2SO_4 or NaOH aliquots. The effect of initial pH on color removal is shown in Fig. 7. It can be noticed that color removal is the most effective (i.e. between 86 and 93.6 %) in the pH range 4.0-7.8. The maximum efficiency of color removal was observed at inlet pH 5.5 with Y_{Col}=93.6%. This corresponds to the region where positively charged $Al(OH)^{2+}$ and $Al_2(OH)_2^+$, and insoluble $Al(OH)_3$ species prevail (Duan and Gregory 2003, Chafi *et al.* 2011): cations are able to conduct primarily charge neutralization, as colloidal particles are usually negatively charged, while insoluble species favor precipitation coagulation. Conversely, Y_{COL} falls when soluble $Al(OH)_4^-$ anions become predominant at high pH.

In Fig. 7, the maximum color removal was observed for an initial pH around 5.5. However, the pH changed during coagulation process, as already described in many papers (Bayramoglu *et al.* 2007, Can *et al.* 2006, Essadki *et al.* 2007, Daneshvar *et al.* 2006, Merzouk *et al.* 2009, Merzouk *et al.* 2009, Cañizares *et al.* 2009, Attour *et al.* 2014). Its evolution depended mainly on the initial pH and the buffering capacities of the liquid to be treated. Indeed, coagulation process exhibits some buffering capacity because of balanced production and consumption of hydroxide anions and presence of Al (III) species. From the values of the final pH values (Table 1) it appears that for initial pH larger than 5 or so pH decreases during coagulation; otherwise, the opposite pH variation is observed. This critical pH value seems to correspond to equilibrium of production and consumption of OH$^-$ anions during the coagulation.

3.4 Effect of inlet dye concentration on the color removal efficiency

Fig. 8 illustrates the variation of color removal with the inlet concentration of the dye, using the optimum conditions obtained previously for mixing rate (60 rpm), mixing time (30 min), coagulant dose (40 mg L^{-1}), settling time (40 min) and pH (5.6). In this figure, dye solutions with different inlet concentrations in the range 25-380 mg L^{-1} were treated by coagulation process. Data show that CC gave satisfactory results for all the concentrations studied, as Y_{COL} was always higher than 80 % and decreased from 91.2 to 82.4% when C_{i} was increased from 230 to 380 mg L^{-1} (Fig. 8). To conclude, the coagulation process based on $Al_2(SO_4)_3$ is robust because it can achieve more than 80 % color removal in a large range of dye concentration. As a matter of fact, the removal yield remains higher than 90 % when C_{i} is in the range 60 - 230 mg L^{-1}. Again, the effect of the settling time remains negligible, particularly when the plateau region of Fig. 8 has been reached.

3.5 Effect of wastewater conductivity on the color removal efficiency

In a general manner, the electrical conductivity affect the thickness of the double layer of any interface in the considered system (reactor wall, electrodes, suspended matter etc.): increasing the conductivity is to reduce this thickness, with possible effect on zeta potential. In electrocoagulation processes, NaCl acts first as supporting electrolyte, but chloride anions may also be oxidized in active chlorine forms, such as hypochlorite anions, that may contribute to pollution removal (Essadki *et al.* 2007). Sodium chloride is usually added in wastewater to increase the ionic strength of the water to be treated, especially when non-ionic pollutants prevail. Indeed, ionic strength reduces zeta potential due to its screening effect on the electrical charges. Increasing water conductivity using NaCl has other advantages: e.g., chloride anions could significantly reduce the adverse effects of other anions, such as HCO_3^-, on coagulation and electrocoagulation (Duan and Gregory 2003, Sánchez Calvo *et al.* 2003). In this work, conductivity and ionic strength of wastewater were adjusted using NaCl in the range 1500-5000 mg L^{-1}. The effect of salt addition on performance of the CC process is shown in Fig. 9.

Experimental results illustrate the weak change in color removal efficiency along time by the electrolyte concentration. The figure shows that Y_{COL} decreased when conductivity was increased. For example, upon κ increase from 830 to 2620 µS cm^{-1}, Y_{COL} was slightly reduced, from 91.8 to 88.1 %. This result, contrary to expectation, results partly from the absence of bicarbonate anions in the synthetic wastewater, while chloride cannot reduce the role of SO_4^{2-} anions as interface co-ordinator with aluminium (Duan and Gregory 2003). As a conclusion, contrary to what has been observed in the treatment by electrocoagulation (Essadki *et al.* 2007, Merzouk *et al.* 2009, Merzouk *et al.* 2009, Chafi *et al.* 2011, Drouiche *et al.* 2012, Adjeroud *et al.* 2015) treatment of the effluent by coagulation is affected by the increase in salinity.

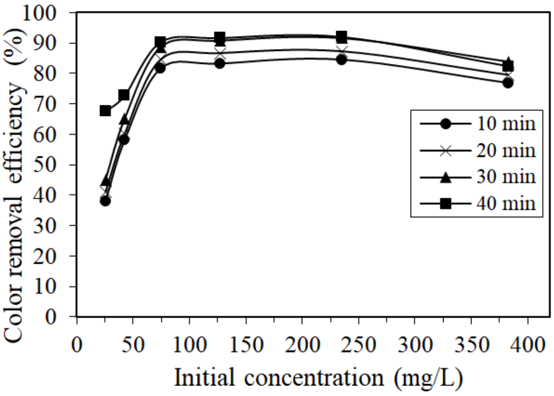

Fig. 8 Effect of initial dye concentration on the color removal efficiency: mixing speed=60 rpm, mixing time=30 min, 40 mg/L of $Al_2(SO_4)_3$, κ=2.4 mS/cm, initial pH 5.6

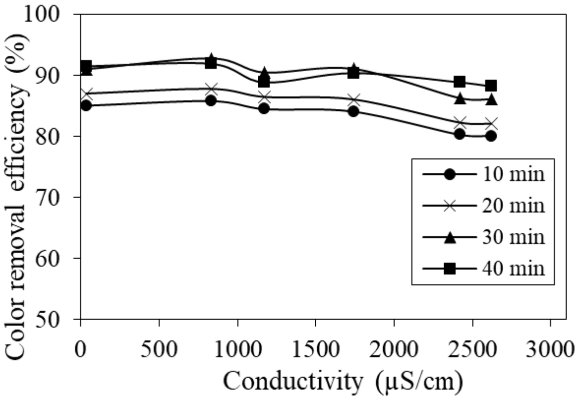

Fig. 9 Effect of salt addition through water conductivity on the color removal efficiency: C_{i}=100 mg/L, mixing speed=60 rpm, mixing time=30 min, 40 mg/L of $Al_2(SO_4)_3$, initial pH 6.5

3.6 Effect of CC process on COD in the effluent stream

The textile wastewater used in this research was taken from the effluent of the textile factory in Algeria (TINDAL-M'sila) which contains the same red dye. The initial COD of the textile wastewater used was about 2500 mg L^{-1}. During the CC process under the following conditions (mixing rate=60 rpm, mixing time=30 min, settling time= 40 min, coagulant dose equal or higher than 40 mg L^{-1}aluminum sulfate, initial pH 8.14 and conductivity κ=5.210 mS cm^{-1}), the COD abatement was 50%. This result shows that the high abatement of the color from the red dye, near 94% does not correspond directly to elimination of the COD because of the colorless compounds present in the effluent.

4. Conclusions

To conclude, the treatment of textile wastewater using the sequential addition of a chemical coagulant enhances clearly color removal and COD reduction and produces lower volume of decanted sludge when aluminum sulfate is used instead of ferric chloride. When the initial pH is near 5, the change in pH during the treatment remains low and coagulation efficiency is maximal with aluminum salts, resulting in 94% color reduction upon addition of coagulant at 40 mg $L^{-1.}$ Additionally, this coagulation procedure is robust, since it can treat efficiently wastewater with dye concentrations up to 380 mg L^{-1} and it depends only slightly on the initial wastewater pH in the range 4.0 and 7.0. Contrary to electrocoagulation, no NaCl addition is required, which can avoid additional environmental issues caused by too high salinity of the water. Finally, other operating parameters such as mixing rate, mixing time and settling time play only a key role when they are higher than the following values: 30 rpm, 20 min and 10 min, respectively. However, contrary to pH value and coagulant dose which mainly depend on physical chemistry of wastewater, an adequate scale-up procedure is required for these parameters before testing the applicability of this coagulation procedure in a continuous wastewater treatment.

References

Adjeroud, N., Dahmoune, F., Merzouk, B., Leclerc, J.P. and Madani, K. (2015), “Improvement of electrocoagulation-electroflotation treatment of effluent by addition of Opuntia ficus indica pad juice”, *Sep. Purif. Technol.*, **144**, 168-176.

Alinsafi, A., Khemis, M., Pons, M.N., Leclerc, J.P., Yaacoubi, A., Benhammou, A. and Nejmeddine, A. (2005), “Electrocoagulation of reactive textile dyes and textile wastewater”, *Chem. Eng. Process.* **44**(4), 461-470.

Amokrane, A., Cornel, C. and Veron, J. (1997), “Landfill leachates pretreatment by coagulation-flocculation”, *Water Res.*, **31**(11), 2775-2782.

Arslan, I.A., Isil, A.B. and Detlef, W.B. (2002), “Advanced oxidation of a reactive dye bath effluent: Comparison of O_3, H_2O_2/UV-C and TiO_2/UV-A processes”, *Water Res.*, **36**(5), 1143-1154.

Attour, A., Touati, M., Tlili, M., Ben Amor, M., Lapicque, F. and Leclerc, J.P. (2014), “Influence of operating parameters on phosphate removal from water by electrocoagulation using aluminum electrodes”, *Sep. Purif. Technol.*, **123**, 124-129.

Barka, N., Qourzal, S., Assabbane, A., Nounah, A. and Ait-Ichou, Y. (2011), “Removal of reactive yellow 84 from aqueous solutions by adsorption onto hydroxyapatite”, *J. Saudi Chem. Soc.*, **15**(3), 263-267.

Bayramoglu, M., Eyvaz, M. and Kobya, M. (2007), “Treatment of the textile wastewater by electrocoagulation: Economical evaluation”, *Chem. Eng. J.*, **128,** 155-161.

Can, O.T., Kobya, M., Demirbas, E. and Bayramoglu, M. (2006), “Treatment of the textile wastewater by combined electrocoagulation”, *Chemosphere*, **62**(2), 181-187.

Cañizares, P., Jiménez, C., Martinez, F., Rodrigo, M.A. and Sáez, C. (2009), “The pH as a key parameter in the choice between coagulation and electrocoagulation for the treatment of wastewaters”, *J. Hazard. Mater.*, **163**(1), 158-164.

Chafi, M., Gourich, B., Essadki, A.H., Vial, C., and Fabregat, A. (2011), “Comparison of electrocoagulation using iron and aluminium electrodes with chemical coagulation for the removal of a highly soluble acid dye”, *Desalination*, **281**, 285-292.

Cook, W.G. and Olive, R.P. (2012), “Pourbaix diagrams for the iron-water system extended to high-subcritical and low-supercritical conditions”, *Corros. Sci.*, **55**, 326-331.

Daneshvar, N., Oladegaragoze, A. and Djafarzadeh, N. (2006), “Decolorization of basic dye solutions by electrocoagulation: An investigation of the effect of operational parameters”, *J. Hazard. Mater.*, **129**(1), 116-122.

Drouiche, N., Aoudj, S., Lounici, H., Drouiche, M., Ouslimane, T. and Ghaffour, N. (2012), “Fluoride removal from pretreated photovoltaic wastewater by electrocoagulation: An investigation of the effect of operational parameters”, *Proc. Eng.*, **33**, 385-391.

Duan, J. and Gregory, J. (2003), “Coagulation by hydrolysing metal salts”, *Adv. Colloid Interfac. Sci.*, **100**, 475-502.

Essadki, A.H., Bennajah, M., Gourich, B., Vial, Ch., Azzi, M. and Delmas, H. (2007), “Electrocoagulation/electroflotation in an external-loop airlift reactor - application to the decolorization of textile dye wastewater: A case study”, *Chem. Eng. Process.*, **47**(8)**,** 1211-1223.

Guida, M., Mattei, M., Rocca, C.D., Melluso, G. and Meriç, S. (2007), “Optimization of alum-coagulation/flocculation for COD and TSS removal from five municipal wastewater”, *Desalination*, **211**(1-3), 113-127.

Holt, P.K., Barton, G.W., Wark, M. and Mitchell, C.A. (2002), “A quantitative comparison between chemical dosing and electrocoagulation”, *Colloid. Surface. A*, **211**(2), 233-248.

Jiang, J.Q. and Graham, J.D. (1996), “Enhanced coagulation using Al/Fe(III) coagulants: Effect of coagulant chemistry on the removal of color-causing NOM”, *Environ. Technol.*, **17**(9), 937-950.

Joo, D.J., Shin, W.S., Choi, J.H., Choi, S.J., Kim, M.C., Han, M.H., Ha, T.W. and Kim, Y.H. (2007), “Decolorization of reactive dyes using inorganic coagulants and synthetic polymer”, *Dyes Pigments*, **73**(1), 59-64.

Joseph, L., Flora, Joseph R.V., Park, Y.G., Badawy, M., Yoon, Y. and Saleh, H. (2012), “Removal of natural organic matter from potential drinking water sources by combined coagulation and adsorption using carbon nanomaterials”, *Sep. Purif. Technol.*, **95**, 64-72.

Kim, S., Park, C., Kim, T.H., Lee, J. and Kim, S.W. (2003), “COD reduction and decolorization of textile effluent using a combined process”, *J. Biosci. Bioeng.*, **95**(1), 102-105.

Kim, T.H., Park, C., Shin, E. and Kim, S. (2002), “Decolorization of disperse and reactive dyes by continuous electrocoagulation process”, *Desalination*, **150**(2), 165-175.

Kim, T.H., Park, C., Shin, E.B. and Kim, S. (2004), “Decolorization of disperse and reactive dye solutions using ferric chloride”, *Desalination*, **161**(1), 49-58.

Kobya, M., Can, O.T. and Bayramoglu, M. (2003), “Treatment of textile wastewaters by electrocoagulation using iron and aluminum electrodes”, *J. Hazard. Mater.*, **100**(1), 163-178.

Lee, J.W., Choil, S.P., Thiruvenkatachari, R. and Shim, W.G. (2006), “Submerged microfiltration membrane coupled with alum coagulation/powdered activated carbon adsorption for complete decolorization of reactive dyes”, *Water Res.*, **40**(3), 435-444.

Lefebvre, E. and Legube, B. (1990), “Coagulation par Fe(III) de substances humiques extraites d'eaux de surface: Effet du pH et de la concentration en substances humiques”, *Water Res.*, **24**(5), 591-606.

Li, W., Hua, T., Zhou, Q., Zhang, S. and Fengxiang, L. (2010), “Treatment of stabilized landfill leachate by the combined process of coagulation/flocculation and powder activated carbon adsorption”, *Desalination*, **264**(1), 56-62.

Lin, S.H. and Peng, C.F. (1994), “Treatment of textile wastewater by electrochemical method”, *Water Res.*, **28** (2), 277-283.

Lin, S.H. and Peng, C.F. (1996), “Continuous treatment of textile wastewater by combined coagulation, electrochemical oxidation and activated sludge”, *Water Res.*, **30**(3), 587-592.

Matilainen, A., Vepsäläinen, M. and Sillanpää, M. (2010), "Natural organic matter removal by coagulation during drinking water treatment: A review", *Adv. Colloid Interfac. Sci.*, **159**(2), 189-197.

Merzouk, B., Gourich, B., Sekki, A., Madani, K., Vial, C. and Barkaoui, M. (2009), "Studies on the decolorization of textile dye wastewater by continuous electrocoagulation process", *Chem. Eng. J.*, **149**(1), 207-214.

Merzouk, B., Gourich, B., Sekki, A., Madani, K. and Chibane, M. (2009), "Removal turbidity and separation of heavy metals using electrocoagulation-electroflotation technique: A case study", *J. Hazard. Mater.*, **164**(1), 215-222.

Mo, J., Hwang, J.E., Jegal, J. and Kim, J. (2007), "Pretreatment of a dyeing wastewater using chemical coagulants, *Dyes Pigments*, **72**(2), 240-245.

Moghaddam, S.S., Moghaddam, M.R. and Arami, M. (2010), "Coagulation/flocculation process for dye removal using sludge from water treatment plant: Optimization through response surface methodology", *J. Hazard. Mater.*, **175**(1), 651-657.

Mollah, M.Y.A., Pathak, S.R., Patil, P.K., Vayuvegula, M., Agrawal, T.S., Gomes, J.A.G., Kesmez, M. and Cocke, D.L. (2004), "Treatment of orange II azo-dye by electrocoagulation (EC) technique in a continuous flow cell using sacrificial iron electrodes", *J. Hazard. Mater.*, **109**(1), 165-171.

Olthof, M. and Eckenfelder, W.W. (1976), "Coagulation of textile wastewater", *Text. Chem. Color.*, **8**(7), 18-22.

Papi´c, S. Koprivanac, N., Boˇzi´c, A.L. and Meteˇs, A. (2004), "Removal of some reactive yes from synthetic wastewater by combined Al(III) coagulation/carbon adsorption process", *Dyes Pigments*, **62**(3), 291-298.

Pela, A. and Tokat, E. (2002), "Color removal from cotton textile industry wastewater in an activated sludge system with various additives", *Water Res.*, **36**(11), 2920-2925.

Rasorio, L.C., Abel, G.E. and Marta, I.L. (2002), "Photo degradation of an azo dye of the textile industry", *Chemosphere*, **48**(4), 393-399.

Rossini, M., Garrido, J.G. and Galluzo, M. (1999), "Optimization of the coagulation-flocculation treatment: Influence of rapid mix parameters, *Water Res.*, **33**(8), 1817-1826.

Sánchez Calvo, L., Leclerc, J.P., Tanguy, G., Cames, M.-C., Paternotte, G., Valentin, G., Rostan, A. and Lapicque, F. (2003), "An electrocoagulation unit for the purification of soluble oil wastes of high COD", *Environ. Prog.*, **22**(1), 57-65.

Selçuk, H., Kaptan, D. and Meriç, S. (2004), "Coagulation of textile wastewater using alum and Fe(III) salts", *Fresen. Environ. Bull.*, **13**(10), 1045-1048.

Semmens, M.J. and Field, T.K. (1980), "Coagulation: Experiences in organics removal", *J. Am. Water Works. Assoc.*, **72**(8), 476-483.

Şengil, I.A. and Özdemir, A. (2012), "Simultaneous decolorization of binary mixture of blue disperse and yellow basic dyes by electrocoagulation", *Des. Water Treat.*, **46** (1-3), 215-226.

Shen, Y.S. and Wang, D.K. (2002), "Development of photoreactor design equation for the treatment of dye wastewater by UV/H_2O_2 process", *J. Hazard. Mater.*, **89**(2), 267-277.

Stanislaw, L., Monika, S. and Renata, Z. (2001), "Biodegradation, decolorization and detoxification of textile wastewater enhanced by advanced oxidation processes", *J. Biotechnol.*, **89**(2), 175-184.

Szygu1a, A., Guibal, E., Palacın, M.A. Ruiz, M. and Sastre, A.M. (2009), "Removal of an anionic dye (Acid Blue 92) by coagulation-flocculation using chitosan", *J. Environ. Manage.*, **90**(10), 2979-2986.

Tahri, N., Masmoudi, G., Ellouze, E., Jrad, A., Drogui, P. and Ben Amar, R. (2012), "Coupling microfiltration and nanofiltration processes for the treatment at source of dyeing-containing effluent", *J. Clean. Prod.*, **33**, 226-235.

Tan, B.H., Teng, T.T. and Mohd Omar, A.K. (2000), "Removal of dyes and industrial dye wastes by magnesium chloride", *Water Res.*, **34**(2), 597-601.

Van der Bruggen, B., Curcio, E. and Drioli, E. (2004), "Process intensification in the textile industry: The role of membrane technology", *J. Environ. Manage.*, **73**(3), 267-274.

Verma, A.K., Bhunia, P. and Dash, R.R. (2014), "Chemical coagulation and sonolysis for total aromatic amines removal from anaerobically pre-treated textile wastewater: A comparative study", *Adv. Environ. Res.*, **3**(4), 293-306.

Verma, A.K., Dash, R.R. and Bhunia, P. (2012), "A review on chemical coagulation/flocculation technologies for removal of colour from textile wastewaters", *J. Environ. Manage.*, **93**(1), 154-168.

Zerrouki, D., Benhadji, A., Taleb Ahmed, M., Djelal, H. and Maachi, R. (2014), "Treatment of a dye solophenyle 4GE by coupling electrocoagulation/nanofiltration", *Membr. Water Treat.*, **5**(4), 251-263.

Zodi, S., Potier, O., Lapicque, F. and Leclerc, J.P. (2010), "Treatment of the industrial wastewaters by electrocoagulation: Optimization of coupled electrochemical and sedimentation processes", *Desalination*, **261**(1), 186-190.

Zodi, S., Potier, O., Michon, C., Poirot, H., Valentin, G., Leclerc, J.P. and Lapicque, F. (2011), "Removal of arsenic and COD from industrial wastewaters by electrocoagulation", *J. Electrochem. Sci. Eng.*, **1**(1), 53-63.

ED