Desalination and Water Treatment
www.deswater.com
doi: 10.5004/dwt.2020.26284

204 (2020) 107–113
November

Electrocoagulation and ozonation: a hybrid technique to treat colored effluents from the textile industry

Gustavo Holz Bracher[a,*], Alejandra Martín-Domínguez[b], Sara Pérez Castrejón[b], Agustín Montes Brito[b], Elvis Carissimi[a]

[a]*Departamento de Engenharia Sanitária e Ambiental, Centro de Tecnologia, Universidade Federal de Santa Maria, Avenida Roraima, 1000, Camobí, 97105-900, Santa Maria, RS, Brazil, Tel. +55 55 3220-8837; emails: gustavohbracher@gmail.com (G.H. Bracher), ecarissimi@gmail.com (E. Carissimi)*
[b]*Coordinación de Calidad del Agua, Instituto Mexicano de Tecnología del Agua, Paseo Cuauhnáhuac, 8532, Col. Progreso, C.P. 62550, Jiutepec, Mor., Mexico, Tel. +52 777 3293-600 (Ext. 505); emails: alejandra.martindominguez@gmail.com (A. Martín-Domínguez), sara_perez@tlaloc.imta.mx (S.P. Castrejón), nitsugamb@gmail.com (A.M. Brito)*

Received 16 June 2019; Accepted 16 June 2020

ABSTRACT

This study evaluated the effectiveness of a hybrid treatment system of ozonation and electrocoagulation for removing color from textile wastewater. The hybrid treatment system was carried out in a semi-pilot scale and continuous flow, with real textile wastewater. Different ozonation and electrocoagulation conditions were simultaneously evaluated in six experimental stages, combining ozone application rates of 0.0 and 13.6 mg/L with the electrical currents of 0.0, 15.8, 22.1 and 28.5 A. The hybrid treatment system had a color removal efficiency of 55.4% after the application of 3.45 g/L of iron and 0.27 g/L of ozone. The increase of electrical current did not increase color removal. Significant color removal efficiencies were only observed in the stages that simultaneously applied both electrocoagulation and ozonation, indicating a synergistic effect. Thus, continuous systems that combine ozonation and electrocoagulation are promising alternatives for future improvements in wastewater treatment from textile industries.

Keywords: Ozonation; Electrocoagulation; Color removal; Textile wastewater; Wastewater treatment; Continuous flow

1. Introduction

Agricultural and industrial developments over the last decades, along with high population growth and urbanization, have aggravated water resource pollution problems. The textile industry is a major industrial sector, and the demand for its products has been increasing; however, its activity is characterized by high water consumption and equally high effluent generation [1–3]. According to Holkar et al. [4], the textile industry is one of the main wastewater generators as a consequence of great water consumption during wet processing operations. In addition, the wastewater generated may present a high pollution potential [4].

The characteristics and composition of textile wastewater vary according to the processes adopted by industry, yet this wastewater generally has high coloration due to the presence of remnant unfixed dyes from the dyeing process [5,6]. Improper discharges of textile wastewater into water bodies can cause various problems for ecosystems, such as blocking the passage of light and inhibiting the biological processes of photosynthesis, as well as presenting risks for living organisms and human health, since some wastewater dyes can be toxic, carcinogenic and mutagenic [3,4,6].

* Corresponding author.

For this reason, color removal from textile wastewater is required.

The removal of color from textile wastewater can be achieved by several wastewater treatment techniques, based on physical, chemical, biological and/or electrochemical processes [7]. Biological degradation processes normally require a large land area and very long treatment times, and are sensitive to effluents with toxic constituents [8,9]. Adsorption processes are also effective for the dissolved colorants from effluents, but they depend on adsorbent regeneration, which involves the loss of expensive absorbent material [10]. Color removal techniques based on membrane separation involve expensive membranes and equipment, produce a concentrate that is hard to dispose of, and show declining performance over time due to membrane fouling [11]. Chemical coagulation can be also used for dye removal, but it generally produces a large amount of sludge [12]. Treatment techniques based on advanced oxidation processes (AOP) have the potential for the treatment of textile wastewater due to their high efficiency of color removal [10]. Ozonation is already a well-established AOP used in full-scale wastewater treatment facilities. It is more energy-efficient than related AOP based on plasma, electrolysis, photocatalysis, ultrasound, and microwaves [13]. Nevertheless, ozonation produces by-products during dye degradation and is not very effective at removing toxic organic compounds [14]. One way to increase the efficiency of the ozonation process for toxic substance removal is to combine it with coagulation processes, which can also increase its efficiency for removing color and organic compounds in wastewater treatment [12]. In this context, the electrocoagulation is an electrochemical process that can replace chemical coagulation. Electrocoagulation can lead to lower sludge production and operational costs and can have higher efficiency for color removal [15]. Batch and laboratory-scale studies have also shown the increase of color removal efficiency with the combination of ozonation and electrocoagulation processes for wastewater treatment, especially using iron electrodes [16–23]. Nevertheless, there is a lack of studies that evaluate the integration of these processes in systems of larger scales and continuous flow with real textile wastewater, which may allow a better view of the application of this integration at the industrial scale.

In this context, the main goal of this study was to analyze the effectiveness of a hybrid treatment system of ozonation and electrocoagulation, on a semi-pilot scale, for the removal of color from textile wastewater, as well as to evaluate the effect of combining these two processes.

2. Material and methods

2.1. Textile wastewater

The wastewater used in this study originated from a textile factory in the state of Puebla, Mexico. The wastewater characteristics are shown in Table 1.

Table 1
Characteristics of textile wastewater used in the study

Parameter	Value
pH	11.2
Electric conductivity (μS/cm)	9,025
Color (ABS_{420nm}/cm)	1.174
Turbidity (NTU)	320
Chemical oxygen demand (mg/L)	3,225

2.2. Treatment system

The wastewater treatment system included a homogenization tank with a working volume of 163.1 L, followed by a 44.2 L ozonation column, an 8.2 L electrocoagulation reactor with iron electrodes, and a 95.5 L settling basin (Fig. 1). This system was designed to work on a semi-pilot scale; it had an overall textile wastewater volume of 312.4 L and workflow of 2.2 L/min. The system had a total residence time of about 1.12 h and was operated with recirculation to study the effect of treatment time. Thus, the settling basin outlet effluent flowed to the homogenization tank to start a new treatment cycle. In the settling basin, the precipitated iron sludge was separated from the liquid effluent, which allowed the removal of color that was probably adsorbed onto the hydroxides formed during electrocoagulation.

2.3. Ozonation system

Ozone was produced by a non-commercial generator, connected to an air compressor (Atlas Copco LFX0.7-10 TM90, Sickla Industriväg 19, Nacka, 105 23 Stockholm, Sweden) which produced an airflow of 1.5 L/min to maintain ozone application in the column at 1.8 g/h (at an ozone concentration of 13.6 mg/L). The ozonation column was made of acrylic and flowed upward, with a diameter of 15 cm and a height of 251 cm. Ozone gas was dispersed in the liquid, at the bottom of the column, by means of a diffuser consisting of a plastic cylinder filled with sand (diameter 8.3 cm, height 9.1 cm). Residual unreacted ozone gas was destroyed by an ozone destroyer equipped with ultraviolet light bulbs.

2.4. Electrocoagulation system

The electrocoagulation system consisted of an acrylic plug flow reactor divided into four modules, each equipped with four sacrificial electrodes (width 15 cm, height 21 cm, thickness 0.5 cm). The electrodes (16 in total) were manufactured from 1,018-grade carbon steel, with 0.3 cm spacing between them, connected in parallel, with a total active surface area of 0.378 m^2.

The electrodes were subjected to galvanostatic direct-current, by means of a power source (Sorensen DCS10-120E, 9250 Brown Deer Road, San Diego, CA, 92121, United States of America). Electrical currents of 15.8, 22.1, and 28.5 A were applied to the system, which corresponded to current densities of 41.8, 58.5, and 75.4 A/m^2. During treatment, polarity was changed every 30 min to avoid the passivation of the electrodes and to promote homogeneous wear. The hydraulic residence time inside the electrocoagulation reactor was 224.7 s, which resulted in flow velocity of 0.020 m/s between the electrodes; this prevented iron accumulation between the electrodes.

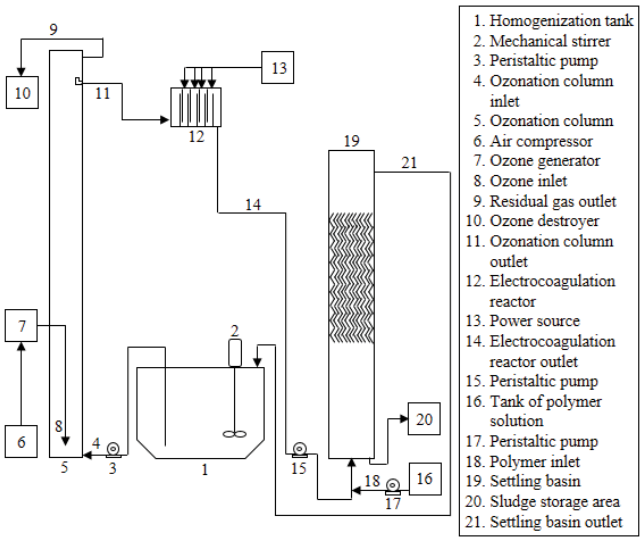

Fig. 1. Treatment system used in the study.

2.5. Settling basin

The settling basin was constructed from acrylic, with a diameter of 25.5 cm and a height of 192.2 cm, which promoted an up-flow velocity of about 0.0007 m/s. The structured packing material was installed in the settling basin in order to promote high-rate sedimentation. The structured packing material was installed at 73.0 cm from the settling basin bottom to a height of 165.2 cm. In the settling basin inlet line, a dose of 1.2 mg/L of anionic polymer SQ673 was added to the electrocoagulation effluent.

2.6. Experimental procedures

During the study, the textile wastewater pH was maintained close to 7.5 through the addition of sulfuric acid, due to the tendency of pH increase during treatment and due to the formation of problematic gel at a lower pH. The study was divided into six experimental stages; wastewater was subject to different treatment conditions at each stage, with a duration time between 10 and 16 h at each one. The treatment conditions in each experimental stage are shown in Table 2.

By means of the different experimental stages, it was possible to separately observe the effects of the electrocoagulation, ozonation, their combination, and the electrical current. For technical feasibility reasons, the stages of the study were executed sequentially, so the initial color intensity in each stage studied corresponded to the final color intensity of the previous stage. Samples were collected from the ozonation column outlet and from the settling basin outlet every 2 h to monitor the variation in effluent color. All the combinations of ozone and the electrical current adopted in the treatment system were determined by preliminary tests with the textile wastewater used in this study, considering the limitations of the equipment used.

Table 2
Treatment conditions for the different experimental stages

Stage	Experimental conditions	
	Ozone applied (mg/L)	Electric current (A)
1	13.6	15.8
2	13.6	22.1
3	13.6	28.5
4	13.6	0.0
5	0.0	0.0
6	0.0	15.8

2.7. Analytical procedures

Color analysis was carried out by absorbance readings in a spectrophotometer (Agilent Cary 60 UV-Vis, G6860A, 5301 Stevens Creek Boulevard, Santa Clara, CA 95051, United States of America), using a quartz cell with a 1 cm optical path. The color was quantified in the wavelength of maximum absorbance in the spectral region of visible light (λ_{max} = 420 nm), which was identified after a spectral scan of each sample between the wavelengths of 190 and 1,100 nm [24,25]. For the analysis, the samples were vacuum-filtered on membranes (MicronSep, pore size 0.45 μm, diameter 47 mm) and subsequently diluted at a ratio of 1:25 with demineralized water.

2.8. Statistical analysis

The effects of ozonation, electrocoagulation and the combination of the processes on wastewater color removal were evaluated for the six experimental stages mentioned above, based on the slopes of color reduction generated from the periodic analysis of the effluent. The comparison between the stages was carried out by analysis of variance (one-way ANOVA) followed by a Tukey test, with a confidence interval of 95%. The stages were considered statistically different when the *p*-value was less than 0.05. To avoid possible distortions and to obtain a more sensitive data analysis, results considered atypical (when the variance of the standardized residual was greater than 2 in each stage) were discarded. All of the statistical analyses were performed using Statistica 7 software.

3. Results and discussion

3.1. Color removal

The results of the wastewater color analysis obtained from samples taken from the ozonation column and from the settling basin, at different times and treatment conditions, are presented in Fig. 2.

Fig. 2 shows a marked decrease in wastewater color during the first three experimental stages, treated by combined ozonation and electrocoagulation. This decrease was not observed in the other experimental stages, when the processes were applied individually (stages 4 and 6) or when only wastewater was recirculated in the system (stage 5).

The greater removal observed in the first three experimental stages demonstrates a synergistic effect. This can be attributed to the formation of reactive oxygen species, such as hydroxyl radicals. Hydroxyl radicals are highly oxidizing agents, which have a rapid kinetic reaction and are not selective, thus they are efficient for the degradation of

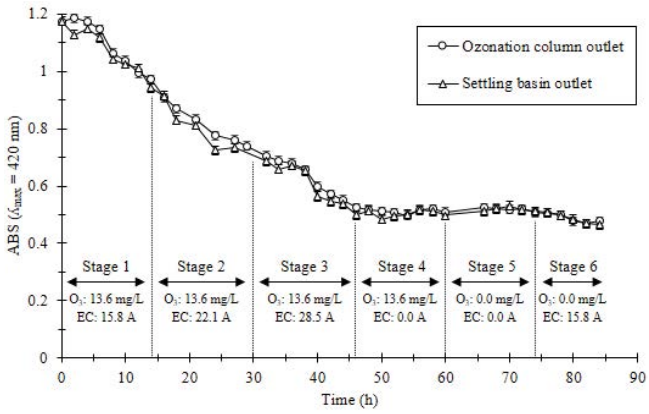

Fig. 2. Kinetics of color removal during different experimental stages.

contaminant compounds, such as dyes [26,27]. Hydroxyl radical formation may be a consequence of the action of Fe^{2+} generated in the electrocoagulation process (Eq. (1)) as a catalytic agent in the decomposition of ozone (Eqs. (2) and (3)). In addition, during ozonation, there can also be the spontaneous formation of hydrogen peroxide (Eqs. (4)–(6)) which can also be converted into hydroxyl radicals from the reaction with Fe^{2+}(Eq. (7)) [21,26,28–30].

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

$$Fe^{2+} + O_3 \rightarrow FeO^{2+} + O_2 \quad (2)$$

$$FeO^{2+} + H_2O \rightarrow Fe^{3+} + {}^{\bullet}OH + OH^- \quad (3)$$

$$O_3 + OH^- \rightarrow O_2 + HO_2^- \quad (4)$$

$$O_3 + HO_2^- \rightarrow HO_2^{\bullet} + O_3^{\bullet-} \quad (5)$$

$$2HO_2^{\bullet} \rightarrow H_2O_2 + O_2 \quad (6)$$

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + {}^{\bullet}OH + OH^- \quad (7)$$

Yet, the synergistic effect observed can be attributed to the oxidation of Fe^{2+} generated in the electrocoagulation process to Fe^{3+}. The ferric constituents, such as ferric hydroxides, generally have less solubility than ferrous constituents and so are capable to promote the formation of larger flocs and more efficient separation of contaminants from the liquid [31–33]. The oxidation of Fe^{2+} to Fe^{3+} may be obtained directly by ozone application in the system (Eqs. (2) and (3)), as well as by the generation of hydrogen peroxide (Eqs. (4)–(7)), oxygen (Eqs. (4)–(7) and (8)–(11)), hydroperoxyl radicals (Eqs. (5) and (13)) and hydroxyl radicals (Eqs. (11) and (14)) in the ozonation process [32,34,35].

$$HO_2^{\bullet} \rightarrow H^+ + O_2^{\bullet-} \quad (8)$$

$$O_2^{\bullet-} + O_3 \rightarrow O_2 + O_3^{\bullet-} \quad (9)$$

$$O_3^{\bullet-} + H^+ \rightarrow HO_3^{\bullet} \quad (10)$$

$$HO_3^{\bullet} \rightarrow OH^{\bullet} + O_2 \quad (11)$$

$$4Fe^{2+} + O_2 + 4H^+ \rightarrow 4Fe^{3+} + 2H_2O \quad (12)$$

$$Fe^{2+} + HO_2^{\bullet} \rightarrow Fe^{3+} + HO_2^- \quad (13)$$

$$Fe^{2+} + HO^{\bullet} \rightarrow Fe^{3+} + OH^- \quad (14)$$

Another possible cause of the synergistic effect may be the breakdown of functional groups and the decrease of organic compounds in the solution, due to oxidation during ozonation. This can cause an alteration in the electrostatic interactions between organic compounds and coagulating agents, and thus increase the affinity of the compounds for the coagulating agent's surface [36]. According to Yan et al. [37], pre-treatment with ozonation can increase the efficiency in coagulation with iron-based coagulants, due to the increase of negatively charged particles, since adsorption is the main mechanism of particle removal by iron coagulants in solutions with a pH higher than 7.5. Barredo-Damas et al. [38] verified positive effects on wastewater from the textile industry, when they included ozonation before coagulation treatments, mainly for organic matter and turbidity removal.

Lin and Liu [39] reported the importance of ozonation for the removal of color from textile wastewater, while chemical coagulation was mainly responsible for the removal of organic compounds and solids from the wastewater. Thus, the absence of significant color removal when only the ozonation process was applied (stage 4) may indicate that the synergistic effect observed was a result of the generation of reactive oxygen species and/or ferric constituents, as previously mentioned.

In the first three stages (Fig. 2), it is also possible to observe a small difference between the color of the samples collected from the ozonation column and the settling basin, at the same treatment times, which indicates the decrease of color promoted by electrocoagulation applied after the ozonation. Fig. 3 shows the decrease of absorbance peak at 420 nm observed at the end of stages 1, 2 and 3, in the different sample collection points. The efficiency of color removal observed after the first three stages were 55.4%, with the application of 3.45 g/L of iron and 0.27 g/L of ozone. The efficiency obtained was higher than that reported by Malik et al. [12] of about 60% of color removal from real textile wastewater with the application of 558.5 g/L of iron and 0.45 g/L of ozone, in a batch treatment system combining ozonation and chemical coagulation. This fact reinforces the possibility of obtaining lower sludge production and operational costs with the use of electrocoagulation instead of the chemical coagulation, due to the higher efficiency of the coagulant consumption in this process. He et al. [16] verified a color removal efficiency of 97% when consuming about 0.2 g/L of iron and 2 g/L of ozone in a batch system, with simultaneous application of electrocoagulation and ozonation, at laboratory-scale. However, He et al. [16] worked with a synthetic solution containing 100 mg/L of a specific azo dye and observed a decline in treatment efficiency as the dye concentration was increased. The authors attributed this decline to the insufficient number of agents to coagulate and oxidize the larger number of molecules at higher concentrations [16]. Thus, the lower color removal with higher consumption of iron observed in this study may be a

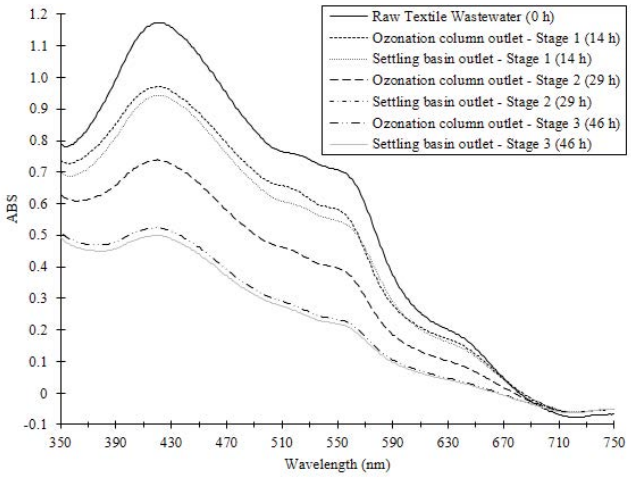

Fig. 3. Spectral scan of textile wastewater before and after the experimental stages 1 (O_3: 13.6 mg/L; EC: 15.8 A), 2 (O_3: 13.6 mg/L; EC: 22.1 A) and 3 (O_3: 13.6 mg/L; EC: 28.5 A).

consequence of the use of real textile wastewater, which may contain a higher dye concentration, as well as a mixture of different dyes and residual constituents from the different processes adopted by the industry, all of which affects treatment performance.

3.2. Statistical analysis

For statistical analysis, only slopes obtained from the ozonation column outlet were used, since the same trend was observed in color removal kinetics at the two sample collection points and less variation was observed in the slopes of the ozonation column outlet. The slopes used in the statistical analysis, after the exclusion of the cases with standardized residual variance greater than 2, are presented in the Table S1.

The results obtained from the statistical analysis reinforce the difference between the kinetics of color removal obtained during the first three experimental stages, in comparison to the others (Table 3). This fact reinforces the hypothesis of the existence of a synergistic effect derived from the combination of ozonation and electrocoagulation processes.

There was no statistically significant difference between stages 4, 5 and 6, which suggests that ozonation and electrocoagulation processes are not effective for wastewater color removal when applied individually under the treatment conditions studied. In addition, no statistically significant difference was observed between stages 1, 2 and 3; this indicates that the increase of electrical current and, therefore, iron ion production in the system, had no effect on wastewater color removal.

It can be proposed that the mechanism of color removal from wastewater was the product of an interaction between ozone and iron ions, proportional to the concentration of both elements in the system, either by the generation of reactive oxygen species, ferric constituents and/or a breakdown of dye compound functional groups, as previously explained [21,37,40]. Thus, the increase of iron in the system had no effect on treatment efficiency (Table 3), possibly because the ozone concentration applied in the system remained constant. So, the increase of iron concentration should be accompanied by an increase in the ozone concentration applied in the system to obtain a higher color removal efficiency.

As no significant statistical difference was observed between color removal slopes for stages 1, 2 and 3, it is possible to determine that the best treatment conditions observed in this study were those applied to stage 1. In this stage, there was less application of electrical current and iron in the treatment system.

From the evaluation of the raw data (Fig. 2), a color removal efficiency of 19.7% was observed in 14 h of treatment for stage 1. Considering the average slope of –0.0108 ABS_{420nm}/h of color removal in the same stage (Table 3), it can be deduced that a treatment time of 108.7 h, corresponding to an application of 5.7 g/L of iron and 0.6 g/L of ozone, should be necessary for the complete removal of wastewater color. This slow slope of color removal may be a consequence of the low ozone concentration applied in the system. Asaithambi et al. [19] obtained 100% color removal from distillery wastewater by ozone assisted electrocoagulation after 4 h of treatment time with a similar concentration of iron (5.6 g/L), but with the use of about thirteen times higher ozone concentration (8 g/L). So, the application of higher ozone concentration, proportional to the iron concentration applied in the electrocoagulation, can promote a higher and faster removal of color in the treatment system

Table 3
Average slopes of color decrease obtained for the different experimental stages

Stage	Ozone applied (mg/L)	Electrical current (A)	Average slope of color removal ± standard deviation (ABS_{420nm}/h)
1	13.6	15.8	–0.0108 ± 0.0038[a]
2	13.6	22.1	–0.0153 ± 0.0044[a]
3	13.6	28.5	–0.0105 ± 0.0036[a]
4	13.6	0.0	–0.0030 ± 0.0026[b]
5	0.0	0.0	–0.0015 ± 0.0027[b]
6	0.0	15.8	–0.0010 ± 0.0035[b]

[a,b]Equal letters indicate values belonging to the same homogeneous group, in which there is no statistically significant difference between its components, according to the Tukey test, at the 95% confidence interval.

studied. It is important to note that the concentrations of iron and ozone were applied slowly for this study and its application in a shorter time could also reduce the treatment time of the system studied.

4. Conclusion

This study evaluated the effect of electrical current and iron concentration applied to the electrocoagulation process, as well as the combination of ozonation and electrocoagulation processes for color removal from the wastewater of a textile factory. Results showed that the integration of ozonation and electrocoagulation processes proved effective for color removal from textile wastewater, with a color removal efficiency of 55.4%. This is in contrast to the individual application of the processes, where no color removal was observed, showing a synergistic effect from this combination. So, the continuous flow systems with the application of ozonation combined with the electrocoagulation are promising alternatives for future improvements in the treatment of wastewater from textile industries. The application of higher ozone concentration in the treatment system presented, proportional to the iron concentration applied, is a possible way of obtaining greater efficiencies of textile wastewater color removal.

Acknowledgments

The authors acknowledge the Mexican Institute for Water Technology (IMTA) for financial support and the Water Quality laboratory team for valuable help in the experimental and analytical procedures. Mr. Gustavo Holz Brächer would like to thank the support received through the Exceed-Swindon Project supported by DAAD (Germany) and CAPES (Brazil) for the scholarship.

References

[1] S. Dey, A. Islam, A review on textile wastewater characterization in Bangladesh, Resour. Environ., 5 (2015) 15–44.
[2] S. Gita, A. Hussan, T.G. Choudhury, Impact of textile dyes waste on aquatic environments and its treatment, Environ. Ecol., 35 (2017) 2349–2353.
[3] S.S. Muthu, Sustainable Fibres and Textiles, Woodhead Publishing, Cambridge, 2017.
[4] C.R. Holkar, A.J. Jadhav, D.V. Pinjari, N.M. Mahamuni, A.B. Pandit, A critical review on textile wastewater treatments: possible approaches, J. Environ. Manage., 182 (2016) 351–366.
[5] A.E. Ghaly, R. Ananthashankar, M.V.V.R. Alhattab, V.V. Ramakrishnan, Production, characterization and treatment of textile effluents: a critical review, J. Chem. Eng. Process. Technol., 5 (2014) 1–18.
[6] T.K.F.S. Freitas, C.A. Almeida, D.D. Manholer, H.C.L. Geraldino, M.T.F. de Souza, J.C. Garcia, Review of Utilization Plant-Based Coagulants as Alternatives to Textile Wastewater Treatment, S.S. Muthu, Ed., Detox Fashion–Waste Water Treatment, Springer, Singapore, 2018, pp. 27–79.
[7] M.C. Collivignarelli, A. Abbà, M.C. Miino, S. Damiani, Treatments for color removal from wastewater: state of the art, J. Environ. Manage., 236 (2019) 727–745.
[8] V. Khandegar, A.K. Saroha, Electrocoagulation for the treatment of textile industry effluent–a review, J. Environ. Manage., 128 (2013) 949–963.
[9] R. Toczyłowska-Mamińska, Limits and perspectives of pulp and paper industry wastewater treatment–a review, Renewable Sustainable Energy Rev., 78 (2017) 764–772.
[10] K. Siddique, M. Rizwan, M.J. Shahid, S. Ali, R. Ahmad, H. Rizvi, Textile Wastewater Treatment Options: A Critical Review, N.A. Anjum, S.S. Gill, N. Tuteja, Eds., Enhancing Cleanup of Environmental Pollutants, Springer, Singapore, 2017, pp. 183–207.
[11] H. Ramlow, R.A.F. Machado, C. Marangoni, Direct contact membrane distillation for textile wastewater treatment: a state of the art review, Water Sci. Technol., 76 (2017) 2565–2579.
[12] S.N. Malik, P.C. Ghosh, A.N. Vaidya, V. Waindeskar, S. Das, S.N. Mudliar, Comparison of coagulation, ozone and ferrate treatment processes for color, COD and toxicity removal from complex textile wastewater, Water Sci. Technol., 76 (2017) 1001–1010.
[13] D.B. Miklos, C. Remy, M. Jekel, K.G. Linden, J.E. Drewes, U. Hübner, Evaluation of advanced oxidation processes for water and wastewater treatment–a critical review, Water Res., 139 (2018) 118–131.
[14] S. Khamparia, D.K. Jaspal, Adsorption in combination with ozonation for the treatment of textile waste water: a critical review, Front. Environ. Sci. Eng., 11 (2017) 1–18.
[15] E. Yuksel, E. Gurbulak, M. Eyvaz, Decolorization of a reactive dye solution and treatment of a textile wastewater by electrocoagulation and chemical coagulation: techno-economic comparison, Environ. Prog. Sustainable Energy, 31 (2012) 524–535.
[16] Z.Q. He, S. Song, J.P. Qiu, J. Yao, X.Y. Cao, Y.Q. Hu, J.M. Chen, Decolorization of CI Reactive yellow 84 in aqueous solution by electrocoagulation enhanced with ozone: influence of operating conditions, Environ. Technol., 28 (2007) 1257–1263.
[17] S. Song, Z.Q. He, J.P. Qiu, L.J. Xu, J.M. Chen, Ozone assisted electrocoagulation for decolorization of CI Reactive black 5 in aqueous solution: an investigation of the effect of operational parameters, Sep. Purif. Technol., 55 (2007) 238–245.
[18] S. Song, J. Yao, Z.Q. He, J.P. Qiu, J.M. Chen, Effect of operational parameters on the decolorization of CI Reactive blue 19 in aqueous solution by ozone-enhanced electrocoagulation, J. Hazard. Mater., 152 (2008) 204–210.
[19] P. Asaithambi, M. Susree, R. Saravanathamizhan, M. Matheswaran, Ozone assisted electrocoagulation for the treatment of distillery effluent, Desalination, 297 (2012) 1–7.
[20] M.A. García-Morales, G. Roa-Morales, C. Barrera-Díaz, V. Martínez Miranda, P. Balderas Hernández, T.B. Pavón Silva, Integrated advanced oxidation process (ozonation) and electrocoagulation treatments for dye removal in denim effluents, Int. J. Electrochem. Sci., 8 (2013) 8752–8763.
[21] J. Behin, N. Farhadian, M. Ahmadi, M. Parvizi, Ozone assisted electrocoagulation in a rectangular internal-loop airlift reactor: application to decolorization of acid dye, J. Water Process Eng., 8 (2015) 171–178.
[22] P. Asaithambi, A.R.A. Aziz, W.M.A.B.W. Daud, Integrated ozone—electrocoagulation process for the removal of pollutant from industrial effluent: optimization through response surface methodology, Chem. Eng. Process.: Process Intensif., 105 (2016) 92–102.
[23] A.R.A. Aziz, P. Asaithambi, W.M.A.B.W. Daud, Combination of electrocoagulation with advanced oxidation processes for the treatment of distillery industrial effluent, Process Saf. Environ. Prot., 99 (2016) 227–235.
[24] M. Nagel-Hassemer, L.A. Coral, F.R. Lapolli, M.T.S.P. Amorim, Processo UV/H_2O_2 como pós-tratamento para remoção de cor e polimento final em efluentes têxteis, Quím. Nova, 35 (2012) 900–904.
[25] M. Punzi, F. Nilsson, A. Anbalagan, B.M. Svensson, K. Jönsson, B. Mattiasson, M. Jonstrup, Combined anaerobic–ozonation process for treatment of textile wastewater: removal of acute toxicity and mutagenicity, J. Hazard. Mater., 292 (2015) 52–60.
[26] A. Mahmoud, R.S. Freire, Métodos emergentes para aumentar a eficiência do ozônio no tratamento de águas contaminadas, Quím. Nova, 30 (2007) 198–205.
[27] T.R.S. Paulino, R.S. Araújo, B.C.B. Salgado, Estudo de oxidação avançada de corantes básicos via reação Fenton (Fe^{2+}/H_2O_2), Eng. Sanit. Ambient, 20 (2015) 347–352.

[28] R.F.P. Nogueira, A.G. Trovó, M.R.A. da Silva, R.D. Villa, M.C. de Oliveira, Fundamentos e aplicações ambientais dos processos Fenton e foto-Fenton, Quím. Nova, 30 (2007) 400–408.
[29] M. Hernández-Ortega, T. Ponziak, C. Barrera-Díaz, M.A. Rodrigo, G. Roa-Morales, B. Bilyeu, Use of a combined electrocoagulation–ozone process as a pre-treatment for industrial wastewater, Desalination, 250 (2010) 144–149.
[30] C.E. Barrera-Díaz, G. Roa-Morales, P.B. Hernández, C.M. Fernandez-Marchante, M.A. Rodrigo, Enhanced electro-coagulation: new approaches to improve the electrochemical process, J. Electrochem. Sci. Eng., 4 (2014) 285–296.
[31] W. Stumm, G.F. Lee, The chemistry of aqueous iron, Schweiz. Z. Hydrol., 22 (1960) 295–319.
[32] A. Bagga, S. Chellam, D.A. Clifford, Evaluation of iron chemical coagulation and electrocoagulation pretreatment for surface water microfiltration, J. Membr. Sci., 309 (2008) 82–93.
[33] M. Vepsäläinen, H. Kivisaari, M. Pulliainen, A. Oikari, M. Sillanpää, Removal of toxic pollutants from pulp mill effluents by electrocoagulation, Sep. Purif. Technol., 81 (2011) 141–150.
[34] W. Stumm, G.F. Lee, Oxygenation of ferrous iron, Ind. Eng. Chem., 53 (1961) 143–146.
[35] L.A. Bernal-Martínez, C. Barrera-Díaz, R. Natividad, M.A. Rodrigo, Effect of the continuous and pulse in situ iron addition onto the performance of an integrated electrochemical–ozone reactor for wastewater treatment, Fuel, 110 (2013) 133–140.
[36] X. Ntampou, A.I. Zouboulis, P. Samaras, Appropriate combination of physico-chemical methods (coagulation/flocculation and ozonation) for the efficient treatment of landfill leachates, Chemosphere, 62 (2006) 722–730.
[37] M.Q. Yan, D.S. Wang, B.Y. Shi, M. Wang, Y. Yan, Effect of pre-ozonation on optimized coagulation of a typical North-China source water, Chemosphere, 69 (2007) 1695–1702.
[38] S. Barredo-Damas, M.I. Iborra-Clar, A. Bes-Pia, M.I. Alcaina-Miranda, J.A. Mendoza-Roca, A. Iborra-Clar, Study of preozonation influence on the physical-chemical treatment of textile wastewater, Desalination, 182 (2005) 267–274.
[39] S.H. Lin, W.Y. Liu, Continuous treatment of textile water by ozonation and coagulation, J. Environ. Eng., 120 (1994) 437–446.
[40] M.-C. Wei, K.-S. Wang, C.-L. Huang, C.-W. Chiang, T.-J. Chang, S.-S. Lee, S.-H. Chang, Improvement of textile dye removal by electrocoagulation with low-cost steel wool cathode reactor, Chem. Eng. J., 192 (2012) 37–44.

Supplemental information

Table S1
Slopes used in the variance analysis and Tukey test, with a standardized residual variance less than two in each stage

Stage	Ozone applied (mg/L)	Electrical current (A)	Number	Slope (ABS_{420nm}/h)
1	13.6	15.8	2	–0.0055
1	13.6	15.8	3	–0.0145
1	13.6	15.8	5	–0.0123
1	13.6	15.8	7	–0.0110
2	13.6	22.1	9	–0.0190
2	13.6	22.1	10	–0.0120
2	13.6	22.1	11	–0.0193
2	13.6	22.1	13	–0.0110
3	13.6	28.5	14	–0.0113
3	13.6	28.5	15	–0.0080
3	13.6	28.5	16	–0.0040
3	13.6	28.5	17	–0.0120
3	13.6	28.5	19	–0.0140
3	13.6	28.5	20	–0.0100
3	13.6	28.5	21	–0.0140
4	13.6	0.0	22	–0.0028
4	13.6	0.0	23	–0.0033
4	13.6	0.0	24	–0.0025
4	13.6	0.0	25	–0.0050
4	13.6	0.0	27	0.0015
4	13.6	0.0	28	–0.0060
5	0.0	0.0	29	0.0004
5	0.0	0.0	30	–0.0005
5	0.0	0.0	31	–0.0020
5	0.0	0.0	32	0.0005
5	0.0	0.0	33	–0.0060
6	0.0	15.8	34	–0.0013
6	0.0	15.8	35	–0.0028
6	0.0	15.8	37	–0.0040
6	0.0	15.8	38	0.0040