

ELECTROCOAGULATION IMPLEMENTATION FOR TEXTILE WASTEWATER TREATMENT PROCESSES

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Abstract Electrocoagulation (EC) has been employed recently to treat tannery, textile, and coloured wastewater. Three main processes are gathered in EC process, namely electrochemistry, coagulation, and flotation. This technique uses DC currents source between metal electrodes immersed in the textile effluent, which causes the dissolution of electrode plates into the effluent. The main advantage of EC compared to chemical coagulation technique is that EC generates less sludge. The objective of the present manuscript is to review the potential of electrocoagulation for the treatment of textile effluent. The most influential factors on removal efficiency, such as initial pH, time of EC, conductivity, current density, initial dye concentration and periodically reversal current on electrodes were discussed. Considering the circular economy concept, which focuses on positive society-wide benefits, manufacturing brick or ceramic materials is feasible method for disposing sludge.

Keywords:

electrocoagulation,
textile effluent,
metal removal,
costs,
current density

1 Introduction

The effluents from textile industry as well as tannery are heavily polluted with heavy metals, such as chromium, and different types of organic matter, especially dyes. The water is dark coloured and does not allow the passage of sun light, therefore, it might be toxic and poorly chemical and biological degradable. (Moussa et al, 2017) Due to toxicity of dyes and metals in textile water, biological treatment is insufficient (Emamjomeh and Sivakumar, 2009).

Chemicals are added to physically remove during traditional techniques such as coagulation or adsorption on traditional adsorbents such as activated carbon in used. In both cases a lot of sludge is generated. Therefore, electrocoagulation (EC) has been employed to treat among other also tannery, textile, and coloured wastewater. The process generates no additional chemicals. In the literature (Moussa et al, 2017) it was stated that more than hundred years electrocoagulation has been applied to remove pollutants from different types of industrial wastewater, such as: emulsion wastewater, coloured wastewater (textile), pulp and paper industry wastewater, tannery, and also from food industry. EC and other electrochemical wastewater treatment processes are considered as an environmentally friendly technology. (Moussa et al, 2017)

Heavy metals, such as Chromium, is very toxic, even carcinogenic as Cr(VI). The Cr removal dynamics was explained (Espinoza et al, 2009). The positive chromium ions are neutralized after movement to the ferrous ions at anode under the electric field, and Cr-ions electrical charge is neutralized. After 15 min, a 93 % drop in Cr concentration was measured as a consequence of the particle coagulation. The electrolysis time of 1 hour was enough for the almost total neutralization of Cr-ions.

The aim of present study was to identify the current state of the EC and review of recent advances in the field of EC. Also ECs' potential as an effective textile wastewater treatment method was considered. From the circular economy concept, disposing sludge could be used as secondary material for manufacturing brick or ceramic materials (Sandoval et al, 2021).

2 Methods

At the iron anode the chemical oxidation to Fe^{2+} take place (Chen, 2004).

Next reaction at pH above 7:



And at pH below 7:



and



From water oxygen and H^+ ions are produced.

At the iron cathode hydrogen is produced:

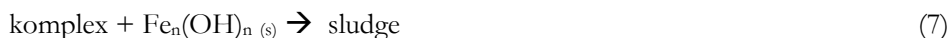


Beside monomeric also polymeric species could form, e.g. $\text{Fe}(\text{H}_2\text{O})_6^{3+}$, $(\text{OH})_2^+$, $\text{Fe}_2(\text{H}_2\text{O})_8(\text{OH})_2^{4+}$ and $\text{Fe}(\text{H}_2\text{O})_6(\text{OH})_4^{4+}$ $\text{Fe}(\text{H}_2\text{O})_5$ if pH changes (Hendaoui et al, 2018).

NaCl increases the production rate of such polymeric species. In textile industry a lot of dyes are present in water. (Eq.5-6) (Nandi and Patel, 2017):



In the second step the adsorption take place, following reactions 7 and 8:



In case of Al, species such as Al^{3+} and $\text{Al}(\text{OH})_2^+$ dominate at low pH. Within the pH range 4 - 9, various species such as $\text{Al}(\text{OH})_2^+$, $\text{Al}(\text{OH})_2^{2+}$, which are monomeric and species such as $\text{Al}_6(\text{OH})_{15}^{3+}$, $\text{Al}_7(\text{OH})_{17}^{4+}$, $\text{Al}_{13}(\text{OH})_{34}^{5+}$ which are polymeric form flocs $\text{Al}(\text{OH})_3(\text{s})$ through complex polymerization and/or precipitation mechanism. When pH is higher than 8, the monomeric $\text{Al}(\text{OH})_4^-$ concentration increases, decreasing the significance of insoluble amorphous $\text{Al}(\text{OH})_3(\text{s})$ flocs (Merzouk et al, 2010). In case of iron electrodes, only soluble Fe^{2+} ions and $\text{Fe}^{3+}(\text{aq})$ ions form the precipitation of $\text{Fe}(\text{OH})_3$ with impurities mainly by charge neutralization or adsorption during coagulation. (Cerqueira, 2009)

Direct red 81 was successfully removed by adsorption (Aoudj et al, 2010). It was observed during analysis of dye-loaded sludge using FTIR. The observed variations in FTIR spectrum suggest the adsorption of dye on $\text{Al}(\text{OH})_3(\text{s})$ flocs.

Some authors emphasized the importance of flotation at EC process (Ghanbari et al, 2014). The sacrificial electrode is electrolytically oxidized and coagulants are formed during an EC process. Such coagulants destabilize the contaminants and consequently agglomerates are formed. As gases are formed according to Eq. 4, pollutants are floated. The process is shown in Fig.1 as one of the main stages involved in EC processes.

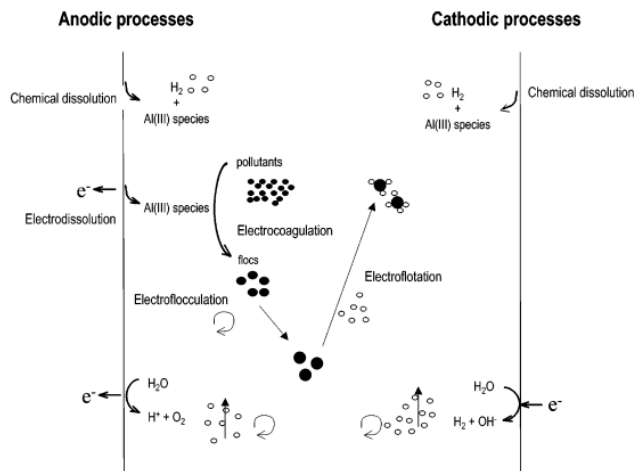


Figure 1: Main mechanism of electrocoagulation

Source: (Canizares et al., 2005)

During EC flocs are formed similar to conventional coagulation. Coloids are neutralised by Al (or Fe) ions and they generate bigger macroflocs which are removed by sedimentation. The stability of colloidal particles is well described by DLVO (Derjaguin-Landua- Verwey-Overbeek) (Moussa et al, 2017). Two forces: attractive van der Waals force with an attraction energy (V_A) and the repulsive electrostatic force with repulsion energy (V_R) are the main forces have the leading role if electrical double-layer is present at particles' surfaces (Fig .2). Around a negative ion an excess of positive ion is accumulated in the interfacial region, and this govern the electrostatic effects. If we sum the Van der Waals attraction energy and electrostatic repulsion energy we gain the net interaction energy of two particles. The total interaction curve ($V_A + V_R$) shows a primary minimum and a secondary minimum in Fig. 2.

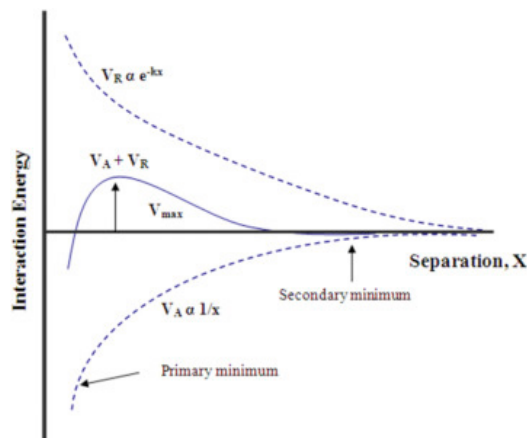


Figure 2: Interaction energy and particle separation curve in dependence of X

Source: (Moussa, 2017)

In general, laboratory EC system consist of 2 electrodes in reactor and are connected to DC source. (Mollah et al 2004).. The research related to the use of electrode made of composite of aluminum and iron which are most widely used electrode material. They both are very concise for the treatment of textile wastewater (Verma, 2017). For real wastewater treatment a cell with two-electrode EC is not appropriate. Large surface area is required. EC cells with monopolar electrodes either in parallel or series connections could improve the efficiency of EC. This arrangement of monopolar electrodes with cells in series is electrically like a single cell. The pair of

»bipolar« electrodes are placed between the two parallel monopolar electrodes and are electrical not connected, there is no interconnections between the anodes. Only the two monopolar electrodes are connected to the electric power source. (Moussa et al, 2017)

3 Effect of important parameters on electrocoagulation efficiency

Since dying is very important branch of industry in developing countries, simple and cost-effective treatment methods are searched for effluent wastewater treatment. EC have potentials for effective method. Authors claim that the most influential factors on removal efficiency are as follows: time of EC, conductivity, pH-value, selected dye concentration, current density, and current on electrodes, which could be periodically reversal (Hakizimana et al, 2017).

3.1 Initial pH value and Conductivity

Neutral pH range was suitable in many experiments, generally at pH 7.0 ± 0.5 . If pH of the dye solutions was between 5.5 and 8.5 almost total colour removal was observed (Daneshvar et al, 2006) and between 6.5 and 10.5 for cationic dye removal (Nandi and Patel, 2017). Initial pH was 7.4 and 70 % of suspended solids was achieved, 97 % of Cr removal and 46 % of COD removal (Apaydin et al, 2009). Best removal results for 5 min electrolysis duration were observed at a pH of 5–9 during the EC process. (Sangil and Ozacar, 2009) and 98 % of reactive dye was degraded. Recent study showed up to 95 % colour removal at pH = 5 and current density 25 mA/cm² (Bener et al., 2019)

The amount of Al(OH)₃ overcomes the amount of OH⁻, then more precipitation will be formed, causing more removal efficiency. (Khoran and Falach, 2018).

The removal of COD and colour from synthetic textile wastewater was studied (Verma, 2014): Up to 86 % colour was removed up to pH=8.

In tannery wastewater the initial pH is around 4 and slightly increases during EC. Up to 85 % of COD could be removed in acid region (Varank et al, 2014).

It could be concluded that pH has great influence on dye removal while the COD removal efficiency was lower, thus, depending on more than one parameter.

Low energy consumption was observed in another study with high Cl⁻ ions content (Nandi Patel). The same study on removal of Brilliant green, which is very well-known cationic dye, showed the influence of NaCl. Dye diffusion and adsorption onto fibre is enhanced by NaCl, whereas Na₂CO₃ is less efficient in dye removal at EC, which is connected to the pH increase.

Almost total colour removal can be obtained in dye solutions with a conductivity of 8 mS/cm. (Daneshvar et al, 2006)

3.2 Reaction time

If reaction time increased 6-fold from 10 min yield in the dye removal increased to 98.3 % (Aoudj et al., 2010). Electrocoagulation time was studied at 7.5 initial pH, 4.0 cm internal electrode distance, and 68 mA cm⁻² current density values. Turbidity was lowered up to 99 %; TSS up to 60 % and Ca-ions up to 80 % in 45 min EC. (Espinoza et al, 2009) Already after 3 min COD decreased by 84 % to 78.5 mg/l with a removal efficiency of when using two iron electrodes with fixed potential of 0.6 V (Zaroual, 2006).

3.3 Current density

The optimum current density of 80 A/m² was used for the colour decrease if dye solution containing BB3 was treated. (Daneshvar et al, 2006) Similar values from 80 to 100 A/m² were reported by Kobya (Kobya et al, 2003). The optimum current density of 75 A/m² was applied for achieving the highest decolourisation of textile wastewater (Ghanbari, 2014). For current density up to 138.9 A/m², more than 90 % brilliant green dye removal was observed after 10 min of operation (Nandi and Patel, 2017). The rate of dye removal increased with increasing of current density. In another study much lower density at 19 A/m² was used and 98 % of dye removal was achieved. (Aoudj et al, 2010)

3.4 Initial dye concentration

The dye concentration is removed by the sufficiency of Fe species. The lower is the dye concentration better would be the removal efficiency. (Zaroual et al, 2006) The same observation was found by Auodj (Aroudi et al, 2010). If dye concentration is high less adsorption sites are available for dye to adsorb.

In general, using a steel anode, the dye decayed in the order: reactive > acid > disperse, and using Al, the order is: acid > reactive > disperse (Garcia-Segura).[22]

3.5 Electrode distance

The interelectrode distance affect differ regarding pollutant nature, hydrodynamic conditions, etc. Best efficiencies were gained at 1.5 cm at 98 % (Aoudj et al, 2010). At such distance, the most of aluminium polymers aggregate in flocs and the dye adsorption was the highest. (Ghanbari et al, 2014). The highest decolorization efficiency was achieved with a distance of 3 cm between the two anodes. At larger distances the flocs interactions are weaker, and the adsorption decreased.

The issue of cathodic polarization also discussed (Wellner et al, 2018) and the results was accumulation of $\text{Al}(\text{OH})_4^-$ at the electrode surface.

3.6 Unit energy demand

The operating costs of EC could be calculated as a sum of the cost of energy, electrode and chemical consumption (Khandegar and Saroha, 2013).

The energy demand for certain dye removal was studied. For 75 % of dye removal the energy demand was 4.7 kWh per kg while it increased to 7.5 kWh per kg if 98 % of dye was removed (Daneshvar et al, 2006) Even lower energy consumption was determined at 0.018 kWh per kg of dye removed (Verma, 2017). 0,59 kWh per kg of dye was needed to achieve colour removal 98 % in textile wastewaters (Ghanbari, 2014) if we assume inlet dye concentration 100 mg L⁻¹ of textile wastewater.

The operation of a continuous photovoltaic electrocoagulation process (PVEC) was investigated, where the photovoltaic module was used instead of the current supply (Khemila, 2018). More than 16 kWh per kg of removed dye were consumed, which was more than by using current supply.

The specific energy demand in relation for aluminium and iron electrodes was studied (Kobyta et al, 2003). Iron electrodes are more energetically efficient than aluminium between pH 5 and 9. The energy consumption was 0.65 kWh/kg COD for iron electrodes, while around 0.8 kWh/kg COD for Al electrodes in acidic medium. In basic mediums costs were twice higher for both types of electrodes. It has been reported that electrocoagulation drastically reduces the cost for the treatment of textile wastewater in comparison with coagulation (Bayramoglu et al, 2007).

4 Conclusion

The industrial potential of electrocoagulation application in textile sector for wastewater treatment was reviewed. The most influential factors were presented and discussed. The pH of the solution is one of the most important operational parameters in EC. As presented in this paper many studies on synthetic textile wastewater were performed using EC as method for COD, dyes, and colour removal. EC seemed to be an efficient process to treat textile wastewater. However, industrial scale up is more difficult as it might seem. Further work is needed to improve the stability of the process especially regarding real textile wastewater treatment, the role of hydrogen gas.

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References

- Moussa D.T., M.H. El-Naas, M. Nasser, M.J. Al-Marri: A comprehensive review of electrocoagulation for water treatment: Potentials and challenges, *Journal of Environmental Management* 186 (2017) 1, 24-41.
- Emamjomeh M. M., M. Sivakumar: Review of pollutants removed by electrocoagulation and electrocoagulation/flotation processes, *J. Environ Manage* 90 (2009) 1663-1679.

- Espinoza-Quiriones F.R., M.M.T. Fornari, A.N. Módenes, S.M. Palácio, F.G. da Silva Jr., N. Szymanski, A.D. Kroumov, D.E.G. Trigueros: Pollutant removal from tannery effluent by electrocoagulation, *Chemical Engineering Journal* 151 (2009), 1-3, 59-65
- Chen G.: Electrochemical technologies in wastewater treatment, *Separation and Purification Technology*, 38 (2004) 1, 11-41
- Hendaoui K., F.Ayari, I. BenRayana, R.BenAmar, F. Darragi, M. Trabelsi-Ayadi: Real indigo *dyeing* effluent decontamination using continuous *electrocoagulation* cell: Study and optimization using Response Surface Methodology, *Process Safety and Environmental Protection* 116 (2018) 578-589
- Nandi B.K., Patel S: Effects of operational parameters on the removal of brilliant green dye from aqueous solutions by electrocoagulation, *Arabian Journal of Chemistry* 10 (2017) S2961-S2968
- Merzouk K., A. Madani: Using electrocoagulation/electroflotation technology to treat synthetic solution and textile wastewater, two case studies, *Desalination*, 250 (2010) 573-577
- Cerqueira A., Russo C., Marques M.R.C: Electroflocculation for textile wastewater treatment, *Brazilian Journal of Chemical Engineering* 26 (2009) 659-668.
- Aoudj S., A. Khelifa, N. Drouiche, M. Hecini, H. Hamitouche: Electrocoagulation process applied to wastewater containing dyes from textile industry, *Chemical Engineering Processing and Process Intensification* 49 (2010), 11, 1176-1182
- Ghanbari E., Moradi M., Eslami A., Emamjomeh M. M.: Electrocoagulation/Flotation of Textile Wastewater with Simultaneous Application of Aluminium and Iron as Anode, *Environmental Processes* 1 (2014) 4, 447-457.
- Cañizares P., M. Carmona, J. Lobato, F. Martínez, M. A. Rodrigo: Electrodeposition of Aluminum Electrodes in Electrocoagulation Processes, *Industrial Engineering Chemical Research* 44 (2005) 12, 4178-4185 (DOI: 10.1021/ie048858a)
- Mollah M.Y.A., Pathak S. R., Patil P.K., Vayuvegula M., Agrawal T.S., Gomes J.A.G., Kesmez M., Cocke D.L.: Treatment of Orange II azo-dye by electrocoagulation (EC) technique in a continuous flow cell using sacrificial iron electrodes, *Journal of Hazardous Materials* 109 (2004) 1-3, 165-171.
- Verma A.K.: Treatment of textile wastewaters by electrocoagulation employing Fe-Al composite electrode, *Journal of Water Process Engineering* 20 (2017) 168-172.
- Hakizimana J. N., Gourich B., Chafi M., Stiriba Y, Vial C., Drogui P., Naja J.: Electrocoagulation process in water treatment: A review of electrocoagulation modeling approaches, *Desalination* 404 (2007) 1-21.
- Daneshvar N., A. Oladegaragoze, N. Djafarzadeh: Decolorization of basic dye solutions by electrocoagulation: an investigation of the effect of operational parameters, *Journal of Hazardous Materials* 129 (2006), 116-122
- Apaydin Ö., U. Kurt, M. Gonullu: An investigation on the treatment of tannery wastewater by electrocoagulation, *Glob. NEST J.* 11 (2009), 4, 546-555
- Şengil, İ.A., M. Özacar: The decolorisation of C. I. Reactive Black 5 in aqueous solution by electrocoagulation using sacrificial iron electrodes, *Journal of Hazardous Materials* 161 (2009) 2-3, 1369-1376.
- Khorrani A. G., Fallah N.: Treatment of textile *dyeing* factory wastewater by *electrocoagulation* with low sludge settling time: Optimization of operating parameters by RSM, *Journal of Environmental Chemical Engineering* 6 (2018)1, 635-642.
- Varank G., H. Erkan, S. Yazıcı, A. Demir, G. Engin: Electrocoagulation of tannery wastewater using monopolar electrodes: process optimization by response surface methodology, *Int. J. Environ. Res.* 8 (2014), 1, 165-180
- Zaroual Z., M. Azzi, N. Saib, E. Chainet: Contribution to the study of electrocoagulation mechanism in basic textile effluent, *Journal of Hazardous Materials* 131 (2006), 1-3, 73-78
- Kobya M., Can, O.T., Bayramoglu, M.: Treatment of textile wastewaters by electrocoagulation using iron and aluminum electrodes, *Journal of Hazardous Materials* 100 (2003) 1-3, 163-178.

- Garcia-Segura S., M.M.S.G. Eiband, J.V. de Melo, C. A. Martinez-Huitl, Electrocoagulation and advanced electrocoagulation processes: A general review about the fundamentals emerging applications and its association with other technologies, *Journal of Electroanalytical Chemistry* 801 (2017) 267-299.
- Wellner D. B., Couperthwaite S. J., G.J. Millar: Influence of operating parameters during electrocoagulation of sodium chloride and sodium bicarbonate solutions using aluminium electrodes, *Journal of Water Process Engineering* 22 (2018) 13-26.
- Khandegar v., Saroha A. K: Electrocoagulation for the treatment of textile industry effluent – A review, *Journal of Environmental Management* 128 (2013) 949-963.
- Khemila B., Merzouk B., Chouder A., Zidelkhir R., Leclerc J.P, Lopicque F.: Removal of a textile dye using photovoltaic *electrocoagulation*, *Sustainable Chemistry and Pharmacy*, 7 (2018) 27-35
- Bayramoglu M., Eyvaz M., Kobya M. Treatment of the textile wastewater by electrocoagulation: economic evaluation, *Chemical Engineering Journal* 128 (2007) 2-3, 155-161
- Can O.T., M. Kobya, E. Demirbas, M. Bayramoglu: Treatment of the textile wastewater by combined electrocoagulation, *Chemosphere*, 62 (2006) 2, 181-187.
- Aleboych A, Daneshvar N, Kasiri M.B.: Optimization of C.I. Acid Red 14 azo dye by electrocoagulation batch process with response surface methodology, *Chemical Engineering and Processing* 47 (2008) 5, 827–832
- Bener S., Bulca O., Palas B., Tekin G., Atalay S., Ersoz G., Electrocoagulation process for the treatment of real textile wastewater: Effect of operative conditions on the organic carbon removal and kinetic study, *Process Safety and Environmental protection* 129 (2019) 47-54.
- Sandoval M.A., Fuentes R., Thiam A., Salazar R : Arsenic and fluoride removal by electrocoagulation process: A general review, *Science of The Total Environment* 753 (2021) 142108.

