



Electrocoagulation for the treatment of textile wastewaters with Al or Fe electrodes: Compared variations of COD levels, turbidity and absorbance

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ABSTRACT

Electrocoagulation technique has been used for the treatment of two wastewaters issued by textile industry. Treatment was carried out in a discontinuous system provided with aluminium or iron electrodes, and with recirculation of the liquid. The efficiency of the technique was followed depending on the electrode material in terms of water treatment, current efficiency of the dissolution, cell voltage, energy consumption to reach the same COD or turbidity abatement: regardless of the quality of the phase separation in the flocculation section downstream of the electrocoagulation cell, the two metals were found to be of comparable efficiency. Besides COD and absorbance were shown to follow similar, regular variations along the treatment; experimental data could be interpreted by a simple model involving the overall equilibrium between the metal dissolved – in the form of hydroxides – and the polluting substance. Abatement of the waste turbidity was observed to obey another law, with a sharp reduction of turbidity after a preliminary phase, where accumulation of metal hydroxide has no effect on this variable.

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1. Introduction

Electrocoagulation is a primary technique for treatment of various wastewaters issued from industry, agriculture or urban areas. The technique relies upon the electrochemical dissolution of sacrificial Al or Fe electrodes. The generated cations contribute to reduce the stability of suspended entities contained by reduction of their zeta potential. Also, upon formation of hydroxides ions at the cathode, metal ions complex to iron or aluminium hydroxides, which are known to be efficient coagulants. The hydrogen bubbles formed at the cathode adsorb the flocs formed by the process, and ensure their flotation, which simplifies their separation from the treated water. The technique has been considered for a number of wastewaters in a very broad range of nature and composition: in particular oil suspensions [1–4], wastes from textile industry [5–7], tannery [8,9] or food processing [10,11]—a list far from exhaustive.

Electrocoagulation has been compared to chemical coagulation for the treatment of liquid wastes, in particular by [12,13]: the small differences of efficiency observed between the two techniques are mainly due to the fact that during the course of the treatment the pH changes in opposite ways: this affects the nature of the coagulant

species formed and the amount of sludge produced. However no general conclusion about the compared efficiency can be made for any type of waste.

Examination of the available literature reveals that the treatment mechanism by electrocoagulation depends on the waste nature, as observed in [14,15] and to some extents by the design of electrocoagulation device and the flow conditions. It also appears that the treatment efficiency has to be considered in terms of various criteria: TOC, COD, turbidity, colour or absorbance at a given wavelength, and concentration of toxic species, e.g. arsenic [4,16] or phosphate [17]. We previously proposed an overall model for the COD abatement of concentrated suspensions, in relation to the removal of turbidity. The present work was aimed at investigating the efficiency of the technique for treatment of textile wastewaters in a discontinuous system with recirculation of the liquid, with special attention to the compared variations of COD levels, turbidity and absorbance.

Besides, the compared efficiency of Fe and Al electrodes has often been investigated and discussed in terms of treatment efficiency and energy consumption [8,14,18,19]. Interestingly, the quality of the water treated and of the sludge formed is also discussed [14,18]. However the conclusions are greatly dependent on the nature of the waste considered. The paper presents the comparison of the two electrode materials for the treatment of textile wastewaters in a discontinuous system, with simultaneous

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variations of COD, absorbance and turbidity. Interpretation of the treatment progress by metal dissolution was also achieved.

2. Experimental

2.1. Chemicals and analytical techniques

Two electrode materials have been used, namely a common mild steel with a C content near 0.3% and very little amounts of other transition metals, and an aluminium alloy with a copper content at 4%. The two materials are respectively referred as iron and aluminium in the paper.

The wastewaters were provided from DMC, a textile industry, St Amarin, France. Two wastewaters have been collected: one directly from the ducts in the factory, the second one collected in the water treatment local station: as a matter of fact the second waste is composed from several wastes emitted by small factories of the area and that are treated in the station by conventional techniques, e.g. precipitation and filtration. The two wastes are labeled “plant” and “station”, depending on their origin. The two effluents were noticeably dyed, with appreciable absorbance in both UV and visible domains. The spectra exhibit comparable profiles in the 250–800 nm, with higher absorption in the low wavelength domain; for the waste collected in the plant a flat absorption peak is observed at 436 nm. Analysis by ion chromatography yielded the concentrations of the most significant anions (Table 1). Conductivity and pH were also measured. The COD levels were determined using the standardized colorimetric method with excess of hexavalent chromium and subsequent measurement of the optical density. Turbidity of the waters was measured using an IR beam at 890 nm and detection of the dispersed light using a Hanna Ins. LP 2000 spectrophotometer. Data were given in Nephelometric Turbidity Unit (NTU). The physicochemical features of the two wastes are reported in Table 1.

Concentrations of Al and Fe in the treated media were determined by atomic absorption (Varian AA-20), after dilution and acidification of the solution samples with nitric acid for total dissolution of the metal species. In most cases, the liquid fractions had to be filtered using conventional 0.45 µm filters to remove the suspended solids, prior to injection into the atomic absorption apparatus.

The density of the two liquids was very close to that of pure water: concentrations are given indifferently in ppm or in mg L⁻¹ in the paper.

2.2. Set-up and protocol

The electrocoagulation cell was a parallel plate electrochemical cell out of methyl polymethacrylate provided with two facing 70 mm × 150 mm electrodes machined in iron or aluminium. In all cases, the two electrodes were of the same material. Experiments were conducted batchwise with recirculation of the liquid in the circuit, which consists of a peristaltic pump, the cell and a double-

walled tank for temperature control at approx. 20 °C and separation of the gas formed. Two litres of wastewater were introduced in the tank and a gentle agitation was ensured along the run. The flow rate of the liquid was fixed at 2.5 cm³ s⁻¹. The current density was fixed in the range 50–200 A m⁻² using an AFX 2930 SB DC power supply and the cell voltage was continuously recorded. Twenty-cubic centimetre samples were taken during the 1-h long runs for assessment of the treatment progress as follows. The pH was observed to increase regularly from its initial value up to 10 within 0.5, depending on the current density applied.

For treatment with Al electrodes, the pH of the samples was adjusted to 6.5 within 0.5 for optimal precipitation of Al hydroxides. For all experiments 1 cm³ of the two-phase samples was taken for analysis of the metal content, the rest was allowed to settle for 24 h. Determination of the COD levels, absorbance at 436 nm and turbidity was made from the clear liquid in the sample vial. All concentrations were corrected from the change in volume due to the regular sampling.

3. Experimental results

3.1. Voltage–current relationship

For the two wastes considered and the two electrode materials, the relationship between the cell voltage and the current density, *j*, has been measured in two ways. First the cell voltage continuously recorded in the galvanostatic electrocoagulation runs, has been averaged over the treatment period. In addition to these runs, the current density in the cell was varied from 10 to 200 A m⁻² in increasing then decreasing scanning as follows. At the considered current density the cell voltage was recorded until steady state was attained, before the current was incremented to its next level. The upward and reverse variations exhibited slightly different profiles, forming a narrow hysteresis loop. This could be due to the change in electrode surface along the experiment and to the increase in solution conductivity by the dissolution of iron or aluminium.

For aluminium non-passivated electrodes, Chen et al. [20] proposed the following relationship:

$$U = -0.76 + \frac{e}{\kappa}j + 0.20 \ln(j) \quad (1)$$

where *e* is the electrode gap. The second term in the right-hand expression corresponds to Ohm's law, the third one being for the overpotentials at the two electrodes. In fact, it was shown that in spite the different redox potentials of the Al/Al³⁺ and Fe/Fe²⁺ couples, near -1.8 and -0.447 V, respectively, the experimental data could be perfectly fitted to the empirical law

$$U = 0.1 + \frac{e}{\kappa}j + 0.20 \ln(j) \quad (2)$$

as exemplified in Fig. 1.

3.2. Efficiency of metal dissolution

Aluminium dissolves according to



Moreover, due to the very negative standard potential of Al/Al³⁺ couple, aluminium can corrode with water in the presence of anions like chloride ions



Besides, iron dissolution is usually considered to form ferrous species as follows:



Table 1

Composition and physicochemical features of the wastewaters considered.

Property	Plant wastewater	Station wastewater
pH	7.06	9.04
Turbidity/NTU	115	110
COD/mg O ₂ L ⁻¹	1787	1237
Conductivity/mS cm ⁻¹	2.8	2.4
[CO ₃ ²⁻]/mg L ⁻¹	2191	1475
[NO ₃ ⁻]/mg L ⁻¹	3.91	11.1
[H ₂ PO ₄ ⁻], [HPO ₄ ²⁻]/mg L ⁻¹	1.9	2.6
[SO ₄ ²⁻]/mg L ⁻¹	144	56
[Cl ⁻]/mg L ⁻¹	84.3	27.9

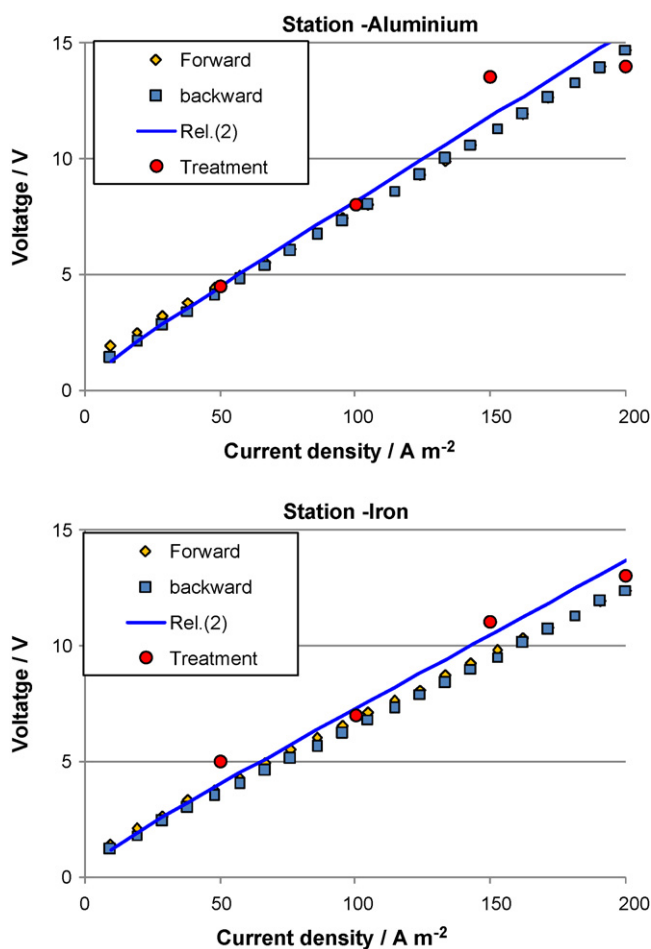


Fig. 1. Variation of the voltage with the current density in the cell for the case of the wastewater collected in the station. Top: with aluminium; bottom, with iron electrodes. "Treatment" refers to wastewater treatment runs.

Ferrous ions precipitate in the form of the $\text{Fe}(\text{OH})_2$ for pH higher than 6. However, ferrous species are unstable in oxygen-containing media and form ferric species by the action of dissolved oxygen



Ferric ions form ferric hydroxides with various hydration states which precipitate into floc of a brownish colour. Although often reported as $\text{Fe}(\text{OH})_3$ in most papers, various oxide-hydroxide species are formed in the electrocoagulation process [14,21].

The current efficiency of metal dissolution was calculated from the amount of metal ion in the liquid sample, m_{metal} , and using Faraday's law

$$\Phi_M = \frac{z_{\text{metal}} F m_{\text{metal}}}{M_{\text{metal}} Q} \quad (7)$$

where subscript "metal" is for Al or Fe, M_{metal} the molecular weight of the metal considered, Q the electrical charge passed and coefficient z_{metal} is equal to 3 for aluminium and 2 for iron, after rels. (3) and (5). The current efficiency was plotted versus the electrical charge (Fig. 2). For the two electrode materials, the efficiency does not vary significantly with time; the observed variations or fluctuations in the first 5–10 min are due to the uncertainty in determination of the low metal concentrations. In spite of a noticeable dispersion, no clear effect of the current density could be evidenced. The average efficiency and the standard deviation were calculated from the experimental data (Fig. 2) and the obtained values are reported in Table 2.

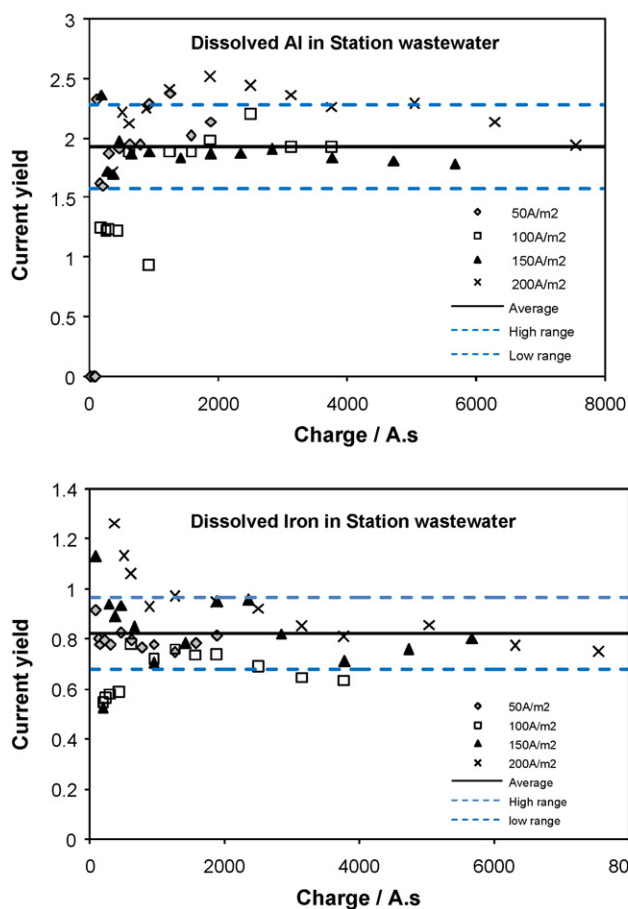


Fig. 2. Current efficiency of aluminium (on top) and iron (bottom) dissolution in the treatment of the wastewater collected in the station.

For aluminium, the current efficiency was found to be near 2.0, indicating the occurrence of side dissolution, which had been formerly evidenced. This side-phenomenon is nevertheless of unexpected significance because of the very low chloride contents in both wastewaters. Iron dissolution occurs quantitatively at the anode surface, as revealed by the high current efficiency larger than 80% (Table 2).

3.3. COD, absorbance and turbidity

COD of the two wastes was shown to decrease regularly along the run from the very first instants, as exemplified by Fig. 3 for the case of the station wastewater. However the COD of the waste could not be totally removed by the treatment, as formerly observed with wastewaters of various nature and origins [2,3,14,15]. With respect to COD, the treatment seems to be at its maximum efficiency for metal ion contents above 500 ppm for Al and 700 ppm for Fe. As shown in Table 3 the remaining COD level after treatment does not depend noticeably on the electrode nature, but more on the origin of the waste: the COD abatement attained 88% for the waste collected in the plant and ranged from 74 to 78% for that from the station.

Table 2
Current yield of metal dissolution in the electrocoagulation cell.

Wastewater	Iron	Aluminium
Plant wastewater	0.80 ± 0.20	2.15 ± 0.38
Station wastewater	0.82 ± 0.14	1.92 ± 0.35

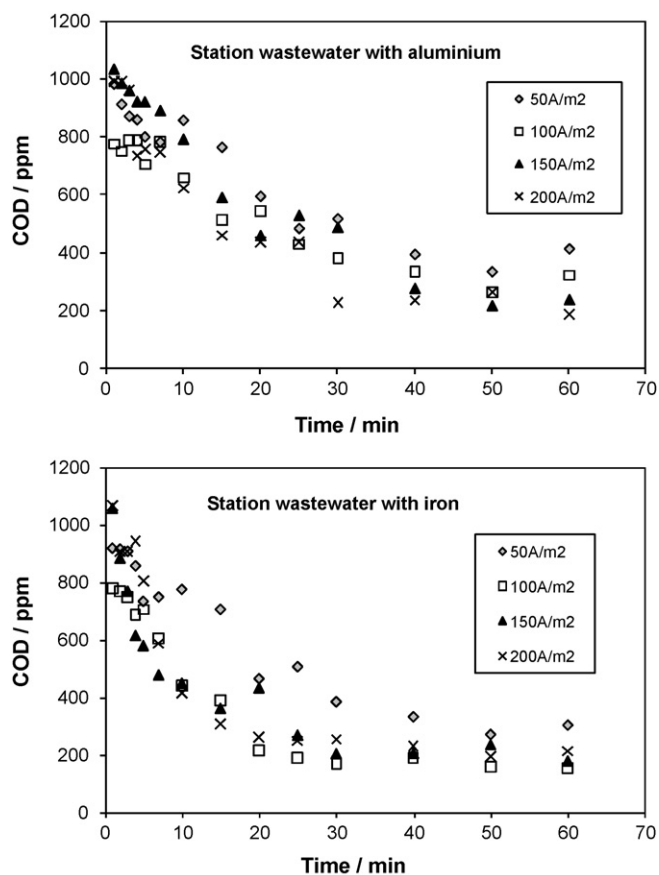


Fig. 3. Time variations of the COD of the Station wastewater in electrocoagulation runs. Top: with aluminium; bottom, with iron electrodes.

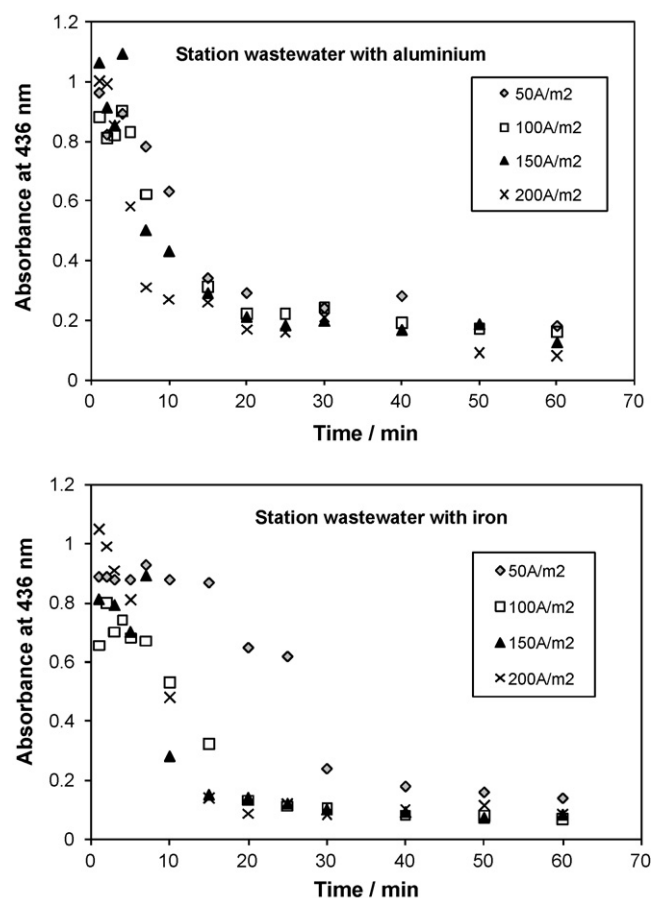


Fig. 4. Time variations of the absorbance (436 nm) of the Station wastewater in electrocoagulation runs. Top: with aluminium; bottom, with iron electrodes.

The absorbance at the 436 nm was observed to follow comparable variations, as exemplified by Fig. 4, with regular reduction of the coloured aspect. Nevertheless the absorbance level tends to zero for amounts of metal dissolved larger than the above values. The remaining COD corresponds to the presence of not visible matter contained in the liquid. The turbidity of the waste was shown to follow a different profile (Fig. 5), with the existence of a steady plateau in the first part of the discontinuous run, followed by a rapid decay down to a zero turbidity level in the final phase of the run. As a matter of fact, turbidity and COD are different variables, expressing different aspects of the waste pollution.

For all experiments, the waste could be faster treated with higher current densities, probably due to faster dissolution of the electrodes. Since the treatment efficiency is to result from the presence of dissolved species, the three observation variables were plotted versus the concentration of metal dissolved, as done in previous papers. In spite of the noticeable deviation around the overall tendency, and because no systematic variation could be observed, the overall plot seems to hold for representation of the experimental variations of COD, turbidity and absorbance at 436 nm in a satisfactory manner, as shown in Fig. 6 for the case of the water collected in the station and treated by using iron electrodes.

Table 3

Maximum COD abatement rate in the electrocoagulation cell.

Wastewater	Iron	Aluminium
Plant wastewater	74%	78%
Station wastewater	88%	88%

4. Interpretation

4.1. Model for COD abatement

Interpretation of the experimental variations was carried out by using an overall model relying upon complexation of the matter suspended by Al species and developed for COD removal of concentrated suspensions [15]. This approach was employed for evaluation of the pollutant complexation by the Al and Fe species. The assumptions of the model are briefly reminded below

- The COD level, $[S]$, expressed in mg O_2 by litre (ppm) varies from $[S]_0$ at initial time, to $[S]_{\text{inf}}$ upon very large amounts of coagulant. The two levels are determined by experiments, depending on the electrode nature and the waste considered.
- Electrocoagulation proceeds by complexation by the dissolved metal species, M



where coefficient n is expressed in mg dissolved metal per mg O_2 .

- Coagulation equilibrium constant was defined by the overall expression [15]

$$K = \frac{[M-S]}{[M]_{\text{free}}[S]} \quad (9)$$

The expression of the COD level can be yielded from mass balances in the metal dissolved and in COD, taking into account rel.

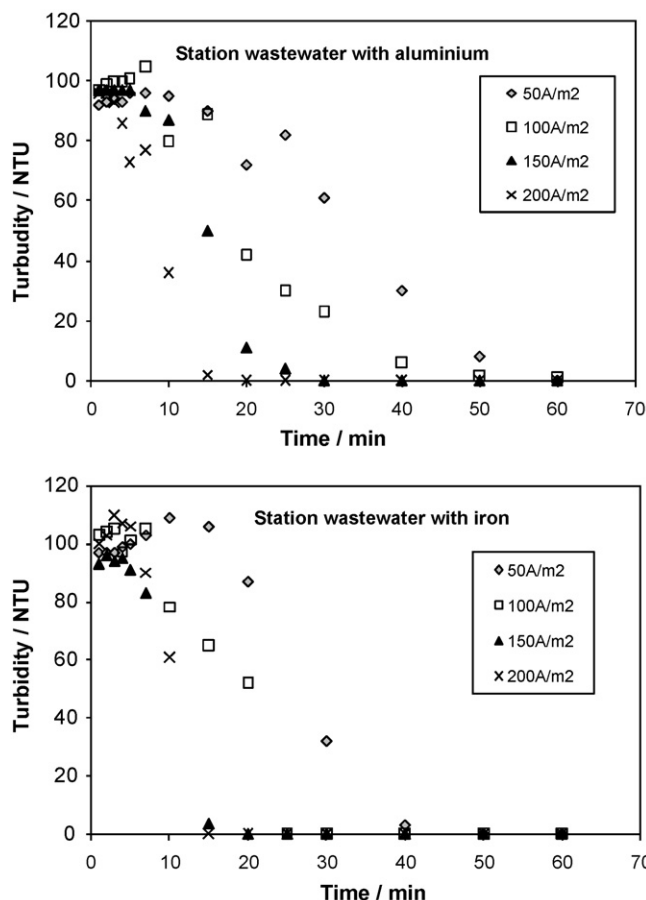


Fig. 5. Time variations of the turbidity of the Station wastewater in electrocoagulation runs. Top: with aluminium; bottom, with iron electrodes.

$$(9) \quad [S] = [S]_{\text{inf}} + \frac{-\left\{1 + K \left(\frac{[M]}{n} - [S]_R\right)\right\} + \sqrt{\left\{1 + K \left(\frac{[M]}{n} - [S]_R\right)\right\}^2 + 4K[S]_R}}{2K} \quad (10)$$

where $[S]_R$ refers to the COD level which can be treated by the coagulation, i.e. the difference $([S]_0 - [S]_{\text{inf}})$.

Fitting of the experimental data to rel. (10) was to yield parameters K and n . However, it was observed that very different couples (K, n) led to similar values of the objective function, calculated as the sum of the squared deviations between experimental and predicted COD levels; for all cases, ratio (K/n) attained comparable values, of the order of $10^{-2} \text{ L mg}^{-1}$. Contrary to the case of oil suspensions with very high COD levels investigated previously [3,15] the present wastewaters have a moderate COD level which can be neglected as compared to $[M]/n$ ratios in rel. (10). Using first-order polynomial expansions, rel. (10) could therefore be simplified to

$$[S] \approx [S]_{\text{inf}} + \frac{[S]_R}{1 + K([M]/n)} \quad (11)$$

For the present case, rels. (10) and (11) yield similar COD variations within 0.2%. Moreover, rel. (11) allows perfect fitting of the experimental data, for the case of the two wastewaters treated by iron or aluminium (Fig. 7). Nevertheless, for the present case of moderate COD levels, only ratio (K/n) could be determined. From rel. (10) it appears that ratio (K/n) is the reciprocal of the amount of metal dissolved required for efficiency water treatment: for example, should the infinite COD levels be neglected, the COD abatement is 90% completed when the global concentration of metal dissolved attains $(9n/K)$. As shown in Table 4, (K/n) is in the range

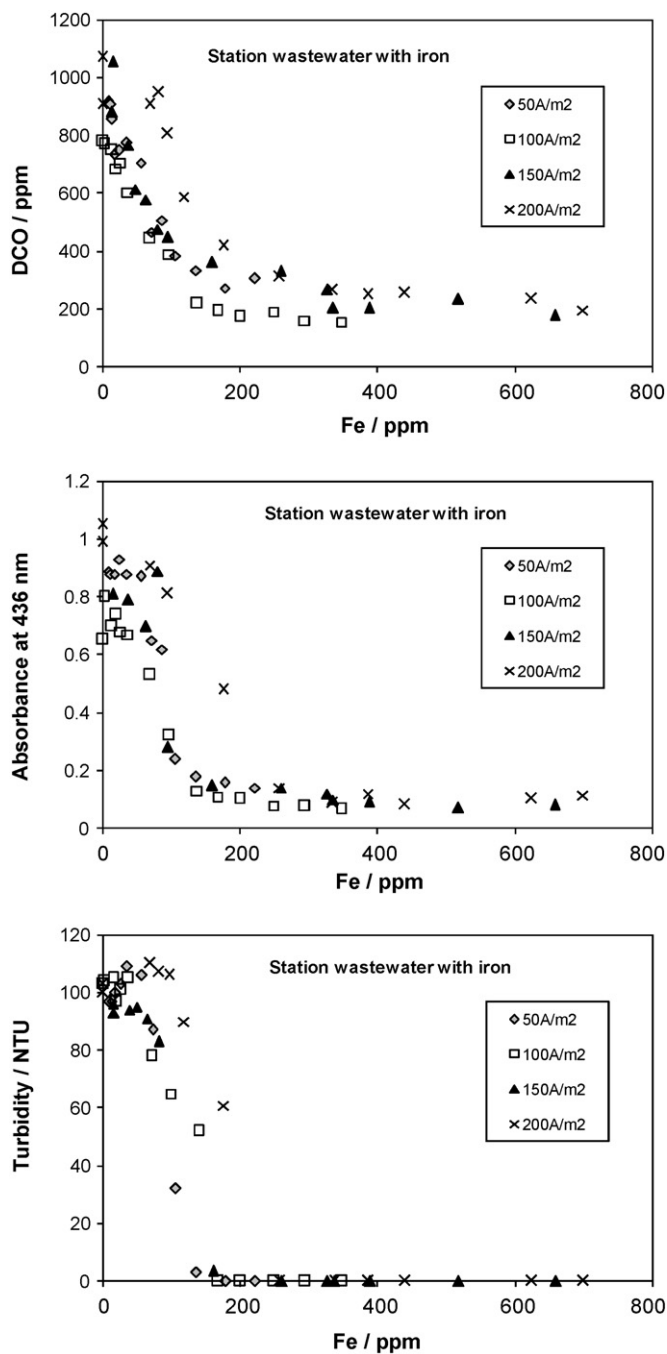


Fig. 6. Variations of the COD, absorbance and turbidity of the Station wastewater with the amount of iron dissolved in electrocoagulation runs.

$0.01\text{--}0.02 \text{ L mg}^{-1}$. Values corresponding to treatment with iron electrodes are nearly 30% higher than those with Al: this is consistent with the experimental observations of higher weight amounts of iron for similar treatment efficiency. Treatment of latex or oil emulsions investigated previously led to K/n ratios ranging from 0.2 to 1, whereas it was found to be near 0.02 for silica suspension, with a far lower COD content.

Table 4
Values of parameter K/n (L mg^{-1}) involved in the approximate model (rel. (11)).

Electrode material	Plant wastewater	Station wastewater
Iron	0.0159	0.0198
Aluminium	0.0099	0.0147

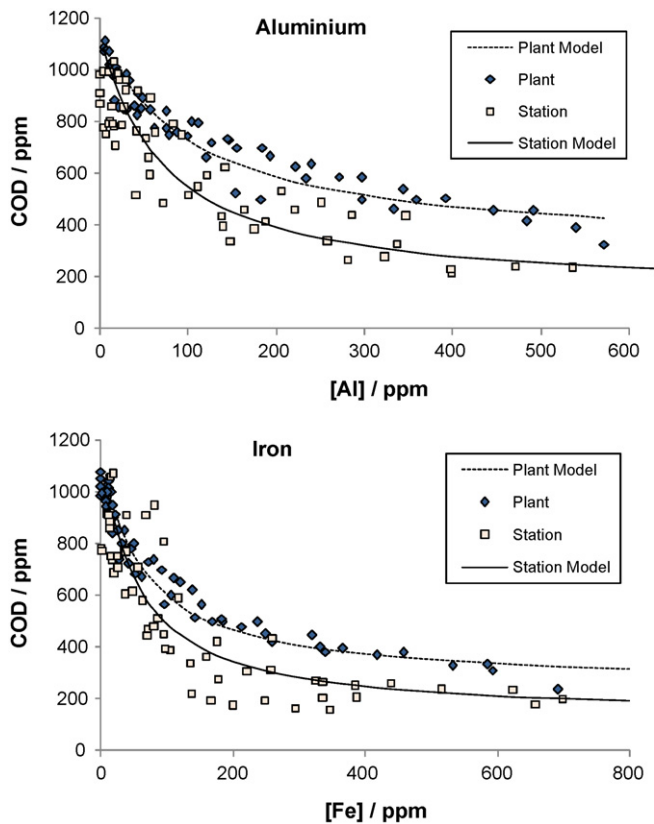


Fig. 7. Variations of the COD with the amount of metal dissolved (top: aluminium; bottom: iron). Comparison of the experimental data to the model prediction (rel. (11)).

4.2. Physical significance of the parameters

COD level $[S]$ varies from approx. 1000 down to 200 mg L⁻¹ in the course of the treatment. Consider the average value of $[S] = 500$ mg L⁻¹ and equivalent concentrations of complexed and non-complexed metals, equilibrium constant K can be evaluated at 2×10^{-3} L mg⁻¹. In addition, the above values for ratio (K/n) lead to n near 0.125 mg Fe/mg COD, and 0.2 mg Al/mg COD.

For the case of organic pollutants, it is often observed that 1 ppm organic carbon corresponds approx. to a COD level of 4 ppm. Taking into account the molecular weights of carbon and of the coagulant metals, it was possible to estimate the number of carbon atoms complexed per atom metal after coagulation. From the results obtained and considering the textile wastewater investigated, one Fe atom would complex around 9 carbon atoms, whereas Al would allow complexation of 3 carbon atoms only. The above estimated “coordination” numbers of Fe or Al largely depend on the wastewater to be treated since one Al atom is involved in the complexation of much more organic matter in the treatment of concentrated oil suspensions [13].

4.3. Compared variations of COD, absorbance and turbidity

The overall model used for prediction of COD abatement along the run was shown to hold in a satisfactory manner for the removal of absorbance at the selected wavelength, as shown in Fig. 8 for the case of the wastewater collected at the treatment station, and for both electrodes: for possible comparison the raw data – variable X – have been normalised after

$$\bar{X} = \frac{X - X_{\text{inf}}}{X_0 - X_{\text{inf}}} \quad (12)$$

Similar conclusion could be made for the other waste (data not shown). As observed by examination of the raw data, absorbance and COD levels are regularly reduced along the run, until their final limits for high amounts of coagulant. On the contrary, the decrease of turbidity does not obey rel. (11). Turbidity remains roughly constant for concentrations of dissolved metals below a threshold, approx. equal to 60 mg L⁻¹ for Al and 100 mg L⁻¹ for Fe. For the two metals, the decrease of the waste turbidity is very steep over this threshold and the turbidity is totally suppressed for metal concentrations larger than 200 mg L⁻¹ for the two coagulants investigated.

The variations described here largely differ from those observed with oils suspensions, for which COD abatement occurs only after a sufficient amount of coagulant is introduced, accompanied by a significant increase in turbidity during this first phase; the reduction of both turbidity and COD had been found to occur rapidly for higher Al amounts. This comparison exemplifies the significant role of the waste nature in the progress of the treatment by electrocoagulation.

4.4. Energy and metal consumed

As shown above, the cell voltage could be modeled by rel. (2). Experiments reported here were carried out with a high electrode gap, resulting in very high values of the cell voltage. For estimation of the energy consumption of a more realistic device, the electrode gap was fixed at 0.3 cm. In spite of the presence of suspended matters in the waste to be treated, electrocoagulation can be successfully carried out with electrode gap varying from 0.3 to 0.5 cm, as shown by previous studies [2,9,17,22]. Consider the treatment

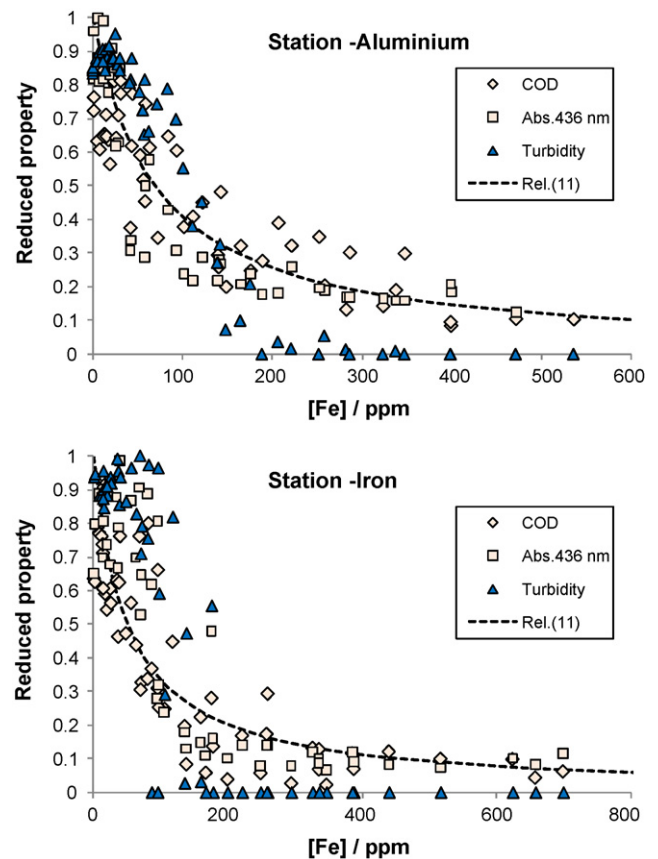


Fig. 8. Reduced COD, absorbance and turbidity (defined by rel. (11)) with the amount of metal dissolved (top: aluminium; bottom: iron). Comparison of the experimental data to the model prediction (rel. (11)).

of a waste with a conductivity at 2 mS cm^{-1} at 100 A m^{-2} : the cell voltage calculated using rel. (2) attains 2.5 V. Besides, the electrical charge required for the waste treatment was calculated considering weight amounts of metal ions at 400 and 600 mg L^{-1} for Al and Fe, respectively. Taking into account the stoichiometry of the metal dissolution, the molecular weight of these metals and the current efficiency of metal dissolution, the energy consumption for treatment of 1000 ppm COD waste was estimated at 1.75 kWh m^{-3} with iron and 1.53 kWh m^{-3} with aluminium. These values could be reduced by 30% by twofold increase in the waste conductivity: addition of sodium chloride at 1 g L^{-1} being of moderate cost and acceptable impact on environment, would allow such a reduction in the energy consumption.

5. Conclusion

Aluminium and iron are suitable electrode materials for the treatment of the investigated textile wastewaters by electrocoagulation. Whereas turbidity and absorbance at 436 nm could be totally removed, COD abatement ranged from 74 to 88% the two wastes considered. The model developed for concentrated suspensions was adapted to the case of wastes with a moderate COD level. Turbidity follows different variations from COD, with a sudden decrease for intermediate concentrations of dissolved metal. Different behaviours of COD and turbidity had been previously observed with concentrated oil suspensions, although with very different profiles along the treatment. The specific response of COD and turbidity in the treatment of wastewaters reveals the difficulty in developing models and theories for the treatment of any waste. The more physical approach developed by Rodrigo's group [23,24] could be promising for this purpose.

The use of the two metals was compared: higher weight amounts of iron have to be dissolved in comparison with aluminium. Taking into account the molecular weights of the metals, the number of electrons exchanged in the anode process together with the current efficiency, treatment by Al or by Fe requires comparable energy consumption. However, use of iron electrodes often results in the formation of very fine brown particles which are less prone to settling than the gel floc formed with aluminium. For further re-use of the treated water, the post-treatment to be added downstream of the electrocoagulation–electroflotation system might represent a penalty to the use of iron over aluminium. This conclusion could also be affected by the efficiency of the solid–liquid separation downstream of the electrolytic cell. Investigations now in progress in the lab with the two coagulating metals would give more precise information on this subject, to be accounted for in the design of a complete treatment process.

Finally the results presented here could be used for economical study of electrocoagulation processes as made previously for the case of soluble oil waste [2] and taking into account the operations required downstream of the electrocoagulation unit.

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