

*Original Article*

# Tertiary treatment of wastewater by electro-coagulation, electro-Fenton and advanced electro-oxidation processes: Comparative and economic study

Faïçal Gaied<sup>1\*</sup>, Boulbaba Louhichi<sup>1</sup>, Mahmoud Bali<sup>2</sup>, and Mohamed Razak Jeday<sup>1</sup><sup>1</sup> *Department of Chemical Process, National School Engineers of Gabes, University of Gabes, Omar Ibn-Elkhattab, Gabes, 6029 Tunisia*<sup>2</sup> *Higher Institute of the Sciences and Techniques of Waters of Gabes, University of Gabes, Cité Zerig, Gabes, 6072 Tunisia*

Received: 13 June 2017; Revised: 14 April 2018; Accepted: 12 June 2018

---

**Abstract**

Due to water scarcity and population growth the use of treated wastewater in agriculture has become more frequent. With this motivation, several technologies have been introduced for the treatment of wastewater. The present work aimed at the tertiary treatment of the polluted water of Tataouine sewage treatment plant (Tunisia) using three alternative processes, namely electro-coagulation (EC), electro-Fenton (EF), and advanced electro-oxidation (AEO). After 180 min, chemical oxygen demand (COD) and biochemical oxygen demand (BOD<sub>5</sub>) abatement rates reached respectively 56% and 14% with sharp deterioration of germs by EC process, and 75% and 52% with complete degradation of germs when EF process was applied. On the other hand, with AEO process, abatement rates of COD and BOD<sub>5</sub> were 92% and 57%, respectively, with complete degradation of germs. The economic study showed that the EF process is technically and economically the best of these alternatives for treatment of this wastewater.

**Keywords:** urban wastewater, microbial loads, organic matter, electro-coagulation, electro-Fenton, advanced oxidation, economic study

---

**1. Introduction**

Water effluents from urban and / or industrial sources are pollutants that are generally laden with organic and mineral matter. These make them aggressive and harmful to the receiving environment if they remain untreated. To overcome the environmental problems that can be caused by these releases and to limit the damage to human health, many treatment processes have been implemented.

Biological treatment is the most widely used technique worldwide, and is mainly used to treat municipal

effluents and industrial biodegradable wastewater. This technique is based on microbial metabolic activity and the ability of microbes to degrade organic matter. The alternative implementations of this type of treatment are activated sludge, biological disc and aerated lagoon (Dinçer & Kargi, 2001; Hammadi *et al.*, 2016; Montalvo *et al.*, 2009; Moura *et al.*, 2009; Palma & Verdonesi, 2009; Wang, Liu, Zhao, Wei, & Sun, 2016; Wu *et al.*, 2016).

Aside from biological techniques there are physico-chemical techniques, which have the ability to treat recalcitrant effluents that are toxic to microorganisms. The removal of pollutants by physicochemical processes can be carried out by flotation, coagulation, sedimentation, filtration, and adsorption. However, these technologies do not completely solve the problem of pollution since they merely displace it, giving rise to sludge and residues that themselves

---

\*Corresponding author

Email address: faicalgaied@gmail.com

require further processing (Baghdadi, Ghaffari, & Aminzadeh, 2016; Bolzonella, Fatone, di Fabio, & Cecchi, 2010; Devi & Saroha, 2017; Dionisi, Bruce & Barraclough, 2014; Hai, Nghiem & Modin, 2013; Hu, Aarts, Shang, Bas, & Rietveld, 2016; Liakos & Lazaridis, 2014; Pitakpoolsil & Hunsom, 2013; Zhang, Liu, Yang, Chen, & Lu, 2014).

The need to develop new wastewater treatment methods that are more efficient and do not generate secondary pollution has become imperative and very pressing. The Anodic Oxidation Process (AEO) has proved to be an effective solution for many bio-resistant or refractory pollutants in synthetic and industrial waters (Brillas & Martínez-Huitle, 2015; Martínez-Huitle, & Ferro, 2006; Martínez-Huitle, Rodrigo, Sires, & Scialdone, 2015; Panizza, & Cerisola, 2009; Sirés, Brillas, Oturan, Rodrigo & Panizza, 2014; Subba Rao & Venkatarangaiah, 2014). These are highly efficient techniques based on the production of highly active radical intermediates, mainly hydroxyl radicals ( $\bullet\text{OH}$ ). Indeed, these very reactive radicals are able to act on the organic matter in a rapid and non-selective manner (Bilińska, Gmurek, & Ledakowicz, 2016; Rodriguez, Santos, & Romero, 2011; Rueda-Márquez *et al.*, 2016; Soares *et al.*, 2016).

These promising techniques have been studied in the treatment of various discharges, such as effluents from textile and agro-food industries. These processes can be used for removing suspended solids, COD, effluent color, oily emulsions, etc (Belaid *et al.*, 2013; Koprivana *et al.*, 2004).

The novelty of this work lies in studying the possibility of using electrochemical techniques for the tertiary treatment of urban wastewater from the treatment plant of Tataouine-Tunisia. The electrochemical treatment was based first on iron electrodes by both electro-coagulation ( $\text{EC}_{\text{iron}}$ ) and electro-Fenton ( $\text{EF}_{\text{iron}}$ ) processes, then on platinum (Pt) electrode by the anodic electro-oxidation process ( $\text{AEO}_{\text{Pt}}$ ) to further reduce the microbial loads and organic matter. Afterwards, an economic evaluation was conducted to study and compare the costs of the studied processes.

## 2. Experimental

The electrochemical treatment was carried out in an electrochemical cell, which consisted of a perfectly stirred discontinuous reactor with two electrodes: iron-iron for  $\text{EC}_{\text{iron}}$  or  $\text{EF}_{\text{iron}}$  and iron-platinum for  $\text{AEO}_{\text{Pt}}$ . The specific surface areas of the electrodes were  $28.5 \text{ cm}^2$  and  $8 \text{ cm}^2$  for iron and platinum electrodes, respectively. The electrode gap was 2 cm. The solution volume was  $0.5 \text{ dm}^3$ . The temperature was kept constant at  $25 \text{ }^\circ\text{C}$ . Magnetic stirring was maintained during the experiments.  $\text{EC}_{\text{iron}}$  and  $\text{EF}_{\text{iron}}$  experiments were carried out under galvanostatic conditions at a current density ( $J_{\text{app}}$ ) of  $57 \text{ mA/cm}^2$ .  $\text{AEO}_{\text{Pt}}$  experiments were conducted at  $250 \text{ mA/cm}^2$ .

The electrodes were inserted into a polytetrafluoroethylene (PTFE) sample holder (Radiometer PEK 29), then they were mechanically eroded using successively finer grades of abrasive paper, polished with  $0.3 \text{ }\mu\text{m}$  alumina, and rinsed with a solution of 1.3 M HCl and then with deionized water.

## 3. Materials and Methods

### 3.1 Quality of inlet and outlet water from the wastewater treatment plant

The inlet and outlet COD and  $\text{BOD}_5$  values for the wastewater treatment plant of Tataouine-Tunisia are given in Table 1.

Table 1. Inlet and outlet COD and  $\text{BOD}_5$  values from the Tataouine-Tunisia wastewater treatment plant.

	Inlet	Outlet
COD ( $\text{mg/dm}^3$ )	860	64
$\text{BOD}_5$ ( $\text{mg/dm}^3$ )	410	21

### 3.2 Measurement of chemical oxygen demand (COD)

COD indicates the amount of organic matter degraded during the process (Lu, Wang, Shan, Li, & Wan, 2006). The COD of different samples at different reaction times was determined by The Standard Methods of American Public Health Association (APHA) (American Public Health Association [APHA], 1999). Two milliliters of each sample were introduced into corresponding digestion tube containing a reagent such as potassium dichromate, sulfuric acid, silver sulphate, or mercury sulphate. These tubes were heated to  $150^\circ\text{C}$  for two hours and then allowed to cool to room temperature. The oxidation took place in the digestion tubes. In fact, the silver salt acts as a catalyst for the oxidation of organic compounds and mercury sulphate saves interferences by chlorides. The COD values were measured using a Photolab VWR S12 analyzer based on the dichromate oxidation method.

### 3.3 Measurement of 5-day biochemical oxygen demand ( $\text{BOD}_5$ )

$\text{BOD}_5$  reflects the amount of biodegradable organic matter contained in water. This biodegradable organic material is evaluated via the oxygen consumed by the microorganisms (Liu & Mattiasson, 2002).

The  $\text{BOD}_5$  of samples taken at predetermined time intervals was determined by the standard method of APHA. The amount of oxygen that is consumed by the microorganisms during biological oxidation of organic solutes over a time period of 5 days is measured (APHA, 1999). The  $\text{BOD}_5$  values were determined by the Lovibond Oxidirect analyzer based on the pressure difference method.

### 3.4 Microbiologic analyses

The objective of the microbiological study was to determine the bacteria concentration remaining after the electrochemical treatment. Microbiological analyses were performed at the Research Veterinary Laboratory in Gabes-

Tunisia. It concerns (GMT) and especially Aerobic bacteria, *Staphylococcus aureus*, *Salmonella-Shigella* (Enterobacteriaceae) and coliform. The culture media for each method are given in Table 2.

### 3.4.1 Aerobic bacteria (GMT)

For this type of bacteria the culture medium used was PCA agar. PCA agar or Glucose agar, which is the yeast extract called by the Anglo-Saxons "Plate Count Agar", is used in food bacteriology for aerobic bacteria enumeration in milk, meats, meat products, or other food products, as well as for the analysis of pharmaceutical products, cosmetics, and their raw materials.

### 3.4.2 *Staphylococcus aureus* (Staph)

For this type of bacteria the culture medium used was the Baird Parker medium. It contains a rich nutrient base and growth accelerators: sodium pyruvate and glycine.

### 3.4.3 *Salmonella* and *Shigella*

For *Salmonella* and *Shigella* the selective medium is *Salmonella-Shigella* Agar (SS Agar) allowing the isolation of pathogenic enteric bacteria (Enterobacteriaceae). For this type of bacteria the culture medium used was Hajna-Kligler, which is a complex medium allowing the search for several biochemical characteristics. It is widely used in Enterobacteriaceae identification.

### 3.4.4 Coliform (*E. coli*)

For this bacteria type, the culture medium used was triphenyltetrazolium chloride (TTC) tergitol. It is mainly used in water colimetry by the filtration method.

## 3.5 The pH measurement

Digital ThermopHmeter (mod BE105, BICASA) equipped with a Metrohm combined glass electrode was used to measure pH of the aqueous solutions. The pH was adjusted using solutions of 0.1 M H<sub>2</sub>SO<sub>4</sub> and/or NaOH.

## 4. Results and Discussion

### 4.1 Treatment of wastewater solutions by electro-coagulation

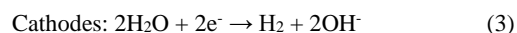
#### 4.1.1 COD degradation

The decrease of the COD in the solution during the electrolysis time at a current intensity (I) equal to 2A is shown in Figure 1. The initial COD (COD<sup>o</sup>) was 64 mg O<sub>2</sub>/dm<sup>3</sup> (Table 1) and decreased during treatment to 30 mg O<sub>2</sub>/dm<sup>3</sup>. This can be explained by the adsorption of organic matter on the insoluble iron hydroxide forms (Fe(OH)<sub>2</sub>, Fe(OH)<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>·xH<sub>2</sub>O), which are known to be strong adsorbents (Brillas, Calpe, & Casado, 2000; Canizares, Jimenez, Martinez, Saez, & Rodrigo, 2007; Lakshmanan, Clifford, & Samanta, 2009). In fact, the compounds Fe(OH)<sub>2</sub> and

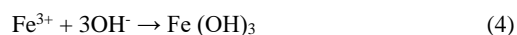
Table 2. Microbiologic analyze for EC<sub>-iron</sub>, EF<sub>-iron</sub> and AEO<sub>-pt</sub> processes.

Time (min)	0	30	60	120	180
Bacteria					
GMT (PCA Agar ) (EC)	21.10 <sup>5</sup>	2.10 <sup>2</sup>	7.10 <sup>2</sup>	15.10 <sup>2</sup>	10 <sup>2</sup>
<i>E.coli</i> (triphenyltetrazolium chloride (TTC) tergitol) (EC)	13.10 <sup>3</sup>	<10	<10	<10	<10
Staph (Baird Parker medium) (EC)	10 <sup>2</sup>	<10	<10	<10	<10
<i>Salmonella</i> (SS Agar) (EC)	<10	<10	<10	<10	<10
GMT (PCA Agar ) (EC) (EF)	29.10 <sup>6</sup>	2.10 <sup>2</sup>	7.10 <sup>2</sup>	5.10	10
<i>E.coli</i> (triphenyltetrazolium chloride (TTC) tergitol) (EF)	13.10 <sup>4</sup>	<10	<10	<10	<10
Staph (Baird Parker medium) (EF)	10 <sup>2</sup>	<10	<10	<10	<10
<i>Salmonella</i> (SS Agar) (EF)	<10	<10	<10	<10	<10
GMT (PCA Agar ) (AEO)	29.10 <sup>6</sup>	2.10 <sup>2</sup>	7.10 <sup>2</sup>	5.10	10
<i>E. coli</i> (triphenyltetrazolium chloride (TTC) tergitol) (AEO)	13.10 <sup>4</sup>	<10	<10	<10	<10
Staph (Baird Parker medium) (AEO)	10 <sup>2</sup>	<10	<10	<10	<10
<i>Salmonella</i> (SS Agar) (AEO)	<10	<10	<10	<10	<10

Fe(OH)<sub>3</sub> are gelatinous metal coagulants generated in the solution during the treatment, which is responsible for the coagulation that enables decantation (Mansouri, Elsaid, Bedoui, Bensalah, & Abdel-Wahab, 2011). In this work, these are generated by dissolution of the iron anode and by water reduction at the iron cathode. Indeed, the main reactions occurring in the electrochemical medium are as follows (1-3):



The resulting Fe<sup>2+</sup> ions are oxidized to Fe<sup>3+</sup>, which combine with the OH<sup>-</sup> formed at the cathode to produce insoluble Fe (OH)<sub>3</sub> as follows (Brillas *et al.*, 2000).



#### 4.1.2 BOD<sub>5</sub> degradation

With EC<sub>iron</sub> process BOD<sub>5</sub> decreased for the first 30 min, and then it remained constant until the end of the treatment (Figure 2). In fact, the BOD<sub>5</sub> decreased from initial 21 mg /dm<sup>3</sup> (Table 1) to 18 mg/dm<sup>3</sup> after 180 min. This can be explained by two facts: the first is due to the adsorption of

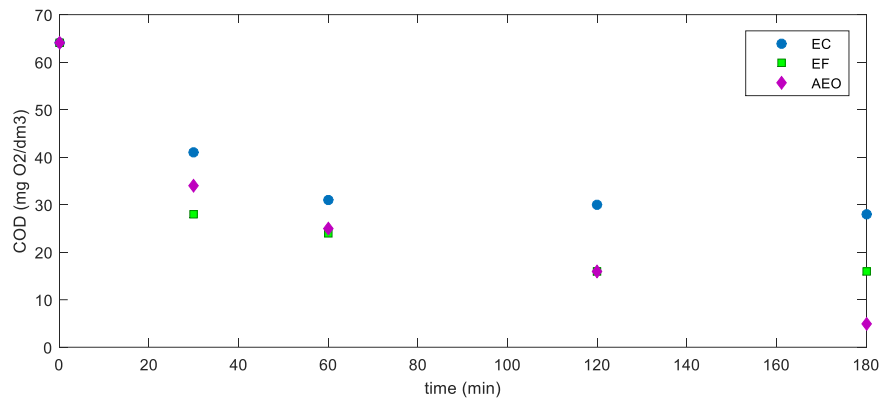


Figure 1. Degradation of COD by EC, EF and AEO processes during the electrolysis time (I=2A, T=25°C).

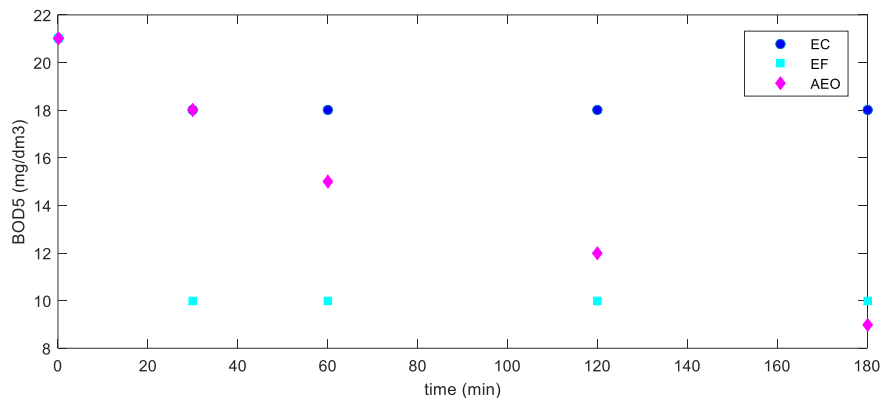


Figure 2. Degradation of DBO<sub>5</sub> by EC, EF and AEO processes during the electrolysis time (I=2A, T=25°C).

organic matter on Fe (OH)<sub>3</sub> mineral monomers and the second one follows from the passage of the current through the wastewater. The latter can be lethal to microorganisms.

#### 4.1.3 pH evolution

Figure 3a shows the pH evolution in the solution versus the electrolysis time for all processes.

As can be seen in Figure 3a, in the case of EC<sub>iron</sub> process, the pH increased towards alkaline and became stable until the electrolysis was completed. This can be explained by the reaction that takes place at the cathode, which produces OH<sup>-</sup>.

### 4.2. Treatment of wastewater solutions by electro-Fenton

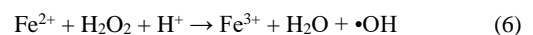
#### 4.2.1 COD degradation

In the EF<sub>iron</sub> process, a reduction of COD from 64 to 16 mg O<sub>2</sub>/dm<sup>3</sup> was observed at pH 3 after a treatment time of 200 min (Figure 1). Fenton processes are more effective in acidic conditions.

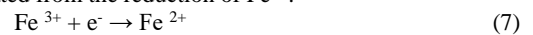
Indeed, the EF<sub>iron</sub> process is based on the Fenton reaction in which H<sub>2</sub>O<sub>2</sub> is generated from the electrochemical reduction of oxygen at the cathode, as follows:



The oxidation power of the generated H<sub>2</sub>O<sub>2</sub> is usually enhanced by the Fe<sup>2+</sup> ions produced by dissolution of iron from the anode to the acid solution (sulfuric acid) to generate free •OH radicals and Fe<sup>3+</sup> ions via the well-known Fenton reaction (Pignatello, Oliveros, & Mackay, 2006):



At the cathode, Fe<sup>2+</sup> ions are continuously regenerated from the reduction of Fe<sup>3+</sup>:



#### 4.2.2 BOD<sub>5</sub> degradation

Figure 2 shows that BOD<sub>5</sub> decreases and then remained constant at 10 mg/dm<sup>3</sup> after 30 min of treatment by the EF process. This may be due to the acidic medium affecting microbes. Moreover, the acidic pH of solution inhibits the development of microbes and causes their mortality.

#### 4.2.3 pH evolution

In EF<sub>iron</sub> process the initial pH must be in the acidic range to generate maximal amounts of •OH and to oxidize the

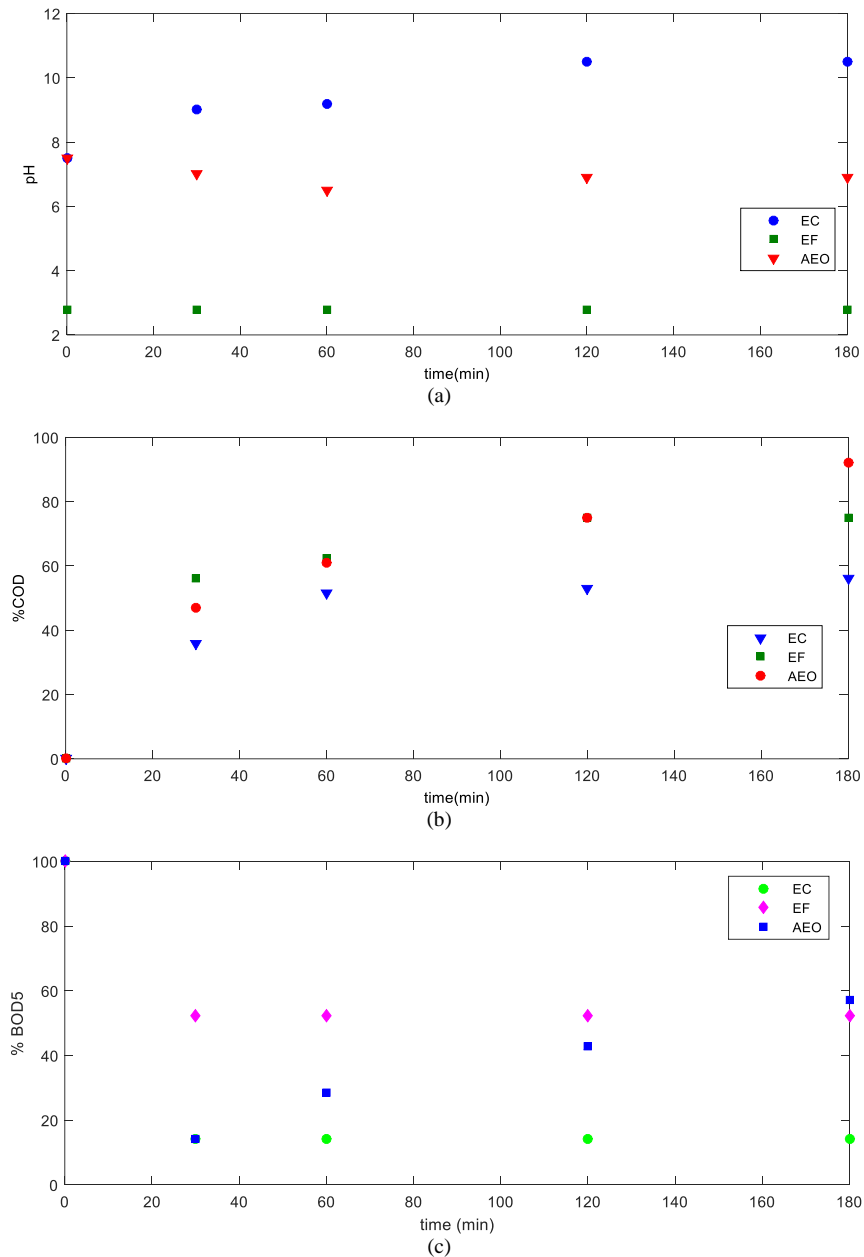


Figure 3. (a) pH evolution, (b) COD removal efficiency, and (c) DBO<sub>5</sub> removal efficiency by EC, EF and AEO processes during the electrolysis time ( $I=2A$ ,  $T=25^{\circ}C$ ).

organic compounds. Most studies have also indicated that the optimum pH for the EF<sub>iron</sub> process is pH 2.8-3, where the catalytic behavior of the  $Fe^{2+}/Fe^{3+}$  coupling can be maintained (Wang, Zheng, Zhang, & Wang, 2016).

### 4.3. Treatment of wastewater solutions by advanced oxidation

#### 4.3.1 COD degradation

As can be seen from Figure 1, the COD decreased progressively during electrolysis reaching  $5mgO_2/dm^3$  at the

end of the treatment. The  $\bullet OH$  radicals, produced directly by the anodic oxidation of water, have the ability to degrade the greatest part of the organic matter in the treated wastewater.

#### 4.3.2 BOD<sub>5</sub> degradation

The BOD<sub>5</sub> decreased to  $9mg/dm^3$  in the AEO<sub>Pt</sub> process (Figure 2). This can be explained by organic matter degradation and bacteria destruction by the actions of  $\bullet OH$  radicals. Also, the current flowing through the wastewater treated can be lethal to these microorganisms.

### 4.3.3 pH evolution

With AEO process, it is observed in Figure 3a that the pH decreased slightly and then became stable. This can be explained by the reaction at the anode, which favors the production of  $\bullet\text{OH}$ . The radicals attack the organic matter and promote the production of short chained carboxylic acids, which acidify the medium, but then the pH increases because of degradation of these acids (Bensalah, Louhichi, & Abdel-Wahab, 2012; Canizares, Paz, Saez, & Rodrigo, 2007; Özcan, Şahin, Koparal, & Oturan, 2007;).

## 4.4. Comparison between EC, EF and AEO

### 4.4.1 COD and BOD removal

After 180 min of electrolysis time, the COD removal efficiency increased to 56%, 75% and 92% (Figure 3c) and the BOD<sub>5</sub> removal efficiency increased to 14%, 52% and 57% (Figure 3b) with EC<sub>iron</sub>, EF<sub>iron</sub> and AEO<sub>Pt</sub> processes, respectively.

The pH, % COD removal and % BOD<sub>5</sub> removal by EC<sub>iron</sub>, EF<sub>iron</sub> and AEO<sub>Pt</sub> processes after 180 min of electrolysis time are given in Table 3. The findings indicate that in AEO<sub>Pt</sub> process the medium remained neutral (pH $\approx$ 7). Therefore, the AEO<sub>Pt</sub> process reached the highest COD and BOD<sub>5</sub> removal.

In EC<sub>iron</sub> process the medium was transformed to alkaline. This can be explained by the generation of great amounts of Fe(OH)<sub>3</sub> and OH<sup>-</sup> that increase pH. The COD and BOD<sub>5</sub> were the lowest among the processes tested. This is may be due to the film, which covered the anode and inhibited the electrolysis. This can reduce the kinetics of organic matter degradation sharply.

Table 3. Removal efficiencies of COD and SEEC by EC<sub>iron</sub>, EF<sub>iron</sub> and AEO<sub>Pt</sub> processes after 180 min.

Process	EC-iron	EF-iron	AEO-pt
pH	10.5	2.8	6.9
COD removal(%)	56	75	92
DBO5 rem(%)	14	52	57

### 4.4.2 Bacteria removal

Table 2 summarizes the microbiological analyses of bacteria (GMT, E. coli, Staph and Salmonella) before and after treatment by EC<sub>iron</sub>, EF<sub>iron</sub> and AEO<sub>Pt</sub> processes. The bacteria counts decreased for all types measured. This may be due to the current passage, which decelerates the micro-organism development in EC<sub>iron</sub> process. However, for EF<sub>iron</sub> and AEO<sub>Pt</sub> treatment, this may be due not only to the effect of current but also to the  $\bullet\text{OH}$  generation, which destroys and kills microorganisms.

## 4.5 Economic evaluation

### 4.5.1 Energy consumption

The energy consumption (W) is necessary to evaluate the energy costs. It was calculated from

$$W (\text{KWh} / \text{m}^3) = Q * U \quad (8)$$

where U is the cell voltage (V) and Q the specific electrical charge (Ah/m<sup>3</sup>).

The specific electrical energy consumption (eq 10) (SEEC, KWh/kgCOD) is calculated from the specific electrical charge (Q) given by equation (9):

$$Q (\text{Ah} / \text{m}^3) = \frac{I \times t}{V} \quad (9)$$

I is current intensity (A), t is time (h) and V is reactor volume (m<sup>3</sup>).

$$\text{SEEC} = \frac{I \times U \times t}{(\text{COD}_{t_0} - \text{COD}_t) \times V} = \frac{Q \times U}{\text{COD}_{t_0} - \text{COD}_t} \quad (10)$$

where COD<sub>t0</sub> = COD<sup>o</sup> and COD<sub>t</sub> (COD at time=t)

Table 4 shows the SEEC values for each studied process, i.e. EC, EF and AEO. The treatment by AEO<sub>Pt</sub> was the most expensive as it consumed the most energy, namely 2.6 kwh/kgCOD among the studied processes, the others being EC<sub>iron</sub> (2.3 kwh/kgCOD) and EF<sub>iron</sub> (2.2 kwh/kgCOD) (Table 4).

Table 4. SEEC for EC<sub>iron</sub>, EF<sub>iron</sub> and AEO<sub>Pt</sub> processes after 180 min.

Process	EC-iron	EF-iron	AEO-pt
SEEC (kwh/kgCOD)	2.3	2.2	2.6

The costs of energy, reagent consumption, iron dissolved and sludge removal are estimated for EC, EF and AEO processes (Tables 5-6). About 0.014 m<sup>3</sup> of sludge generated / m<sup>3</sup> of solution was determined experimentally for the EC and EF processes. No deposit of sludge was detected during the treatment by EAOP<sub>t</sub> because the platinum anode is dimensionally stable. The unit electricity price for industrial use is of the order 0.14€ per KWh and the total costs of 1 m<sup>3</sup> of treated wastewater is in the range commonly seen in the literature; and the cost of sludge disposal is assumed to be 150 € m<sup>-3</sup> (157.868 \$ m<sup>-3</sup>) (Canizares, Paz, Saez, & Rodrigo, 2009).

The price of the main equipment is given by the William equation (Llanos, Camarillo, Perez, Canizares, & Rodrigo, 2011). This estimate assumes 10-year linear type, without value for money update. In fact, the cost P of the electrochemical reactor estimated in 2010 is

$$P = 21662 A^{0.7953} \quad (11)$$

Here A is the electrode area (Cañizares *et al.*, 2007):

$$A = \frac{Q(\text{Ah/m}^3) \times 1000 \times \text{flow rate} (\text{m}^3/\text{d})}{j_{\text{app}} (\text{A/m}^2) \times 24} \quad (12)$$

As shown in Table 7 the reactor cost and total capital investment (without anode, for flow rate 1 m<sup>3</sup>.d<sup>-1</sup>) (in €) was estimated to be 19518.3€ and 111059.1€, respectively for EC<sub>iron</sub> and EF<sub>iron</sub>. However, the estimates for AEO<sub>Pt</sub> were 6022.9€ for reactor cost and 34270.5€ for total capital

Table 5. Estimated cost per 1 m<sup>3</sup> wastewater (COD<sup>o</sup>=64mgO<sub>2</sub>/dm<sup>3</sup>) of reagents used in each process, of iron dissolved, and of sludge generated by both EC<sub>-iron</sub> and EF<sub>-iron</sub>.

Process	EC <sub>-iron</sub>	EF <sub>-iron</sub>	AEO <sub>-pt</sub>
Reagent added	H <sub>2</sub> SO <sub>4</sub>	H <sub>2</sub> SO <sub>4</sub>	H <sub>2</sub> SO <sub>4</sub>
Quantity added (kg/m <sup>3</sup> ) solution	0	0.2	0
Reactive cost (€)	0.52	0.52	0.52
Total cost of reagents added (€)	0	0.104	0
amount of anode matter dissolved(kg/m <sup>3</sup> )	2	2	0
cost of iron anode matter dissolved(€)	1.001	1.001	1.001
cost of sludge treatment (€/m <sup>3</sup> )	150	150	150
Amount of sludge (m <sup>3</sup> ) (0.014 m <sup>3</sup> sludge/ m <sup>3</sup> )	0.014	0.014	0
cost of sludge treated (€)	2.10	2.10	0

Table 6. Quantity of energy consumed (W). Estimated energy cost and total cost (anode price not included) of each process. (Electricity price 0.14 € / kwh used in estimates.)

Process	EC <sub>-iron</sub>			EF <sub>-iron</sub>			AEO <sub>-pt</sub>		
Time (min)	60	120	180	60	120	180	60	120	180
W (KWh/m <sup>3</sup> )	28	56	84	36	72	108	52	104	156
Energetic cost (€)	3.9	7.8	11.7	5.1	10.1	15.1	7.3	14.6	21.8
cost of sludge treated (€)	2.10	2.10	2.10	2.10	2.10	2.10	0	0	0
Total cost of reagents added (€)	0	0	0	0.104	0.104	0.104	0	0	0
Total cost (€)	6.0	9.9	13.8	7.3	12.3	17.3	7.3	14.6	21.8

Table 7. Reactor cost and total capital investment (in €) of EC<sub>-iron</sub>, EF<sub>-iron</sub> and AEO<sub>-pt</sub> (without anode, flow rate 1m<sup>3</sup>.d<sup>-1</sup>).

	Q (Ah.m <sup>-3</sup> )	A (m <sup>2</sup> )	P (€)	Total capital investment(€)
EC <sub>-iron</sub> or EF <sub>-iron</sub>	12	0.877	19518.3	111059.1
AEO <sub>-pt</sub>	12	0.2	6022.9	34270.5

investment. In this work,  $j_{app}(AEO_{Pt}) \geq j_{app}(EC_{iron} \text{ or } EF_{iron})$  so  $A(AEO_{Pt}) \leq A(EC_{iron} \text{ or } EF_{iron})$ . Consequently, the AEO<sub>Pt</sub> process had the lowest reactor cost but required the use of a platinum electrode, which is very expensive and rare. The treatment cost of 1 m<sup>3</sup> of wastewater was estimated at 13.8, 17.3 and 21.8 €/m<sup>3</sup> respectively for EC<sub>iron</sub>, EF<sub>iron</sub> and AEO<sub>Pt</sub> processes. This is due to the availability of iron and it being less expensive than platinum.

## 5. Conclusions

In the present work we conducted tertiary treatment experiments on wastewater and showed that:

- COD removal efficiency increased in the treated wastewater by EC<sub>iron</sub>, EF<sub>iron</sub> and AEO<sub>Pt</sub> processes respectively up to 56%, 75% and 92%.
- The BOD<sub>5</sub> removal efficiency reached 14%, 52% and 57% with EC<sub>iron</sub>, EF<sub>iron</sub> and AEO<sub>Pt</sub> processes, respectively.
- The bacteria counts decreased for the studied types (GMT, E. coli, Stoph and Salmonella) after treatment by EC<sub>iron</sub>, EF<sub>iron</sub> and AEO<sub>Pt</sub> processes. This is due to the current passage, which decelerates microorganism development and destroys them.

- The treatment cost of 1 m<sup>3</sup> of wastewater was estimated at 13.8, 17.3 and 21.8 €/m<sup>3</sup> for EC<sub>iron</sub>, EF<sub>iron</sub> and AEO<sub>Pt</sub> processes respectively.

- Reactor cost and total capital investment (without anode, for flow rate 1 m<sup>3</sup>.d<sup>-1</sup>) (in €) were 19518.3 € and 111059.1 € respectively for both EC<sub>iron</sub> and EF<sub>iron</sub>. However, estimates for AEO<sub>Pt</sub> were 6022.9€ for reactor cost and 34270.5€ for total capital investment.

Wastewater treatment by the different processes reduced organic and bacterial loads. These waters could then be used for irrigation of some types of plants, such as forages. An economic study of amortization is required for proper choice of the best alternative process.

## Acknowledgements

This work was supported by the Ministry of the Higher Education and Scientific Research in Tunisia.

## References

- American Public Health Association. (1999). *Standard methods for the examination of water and wastewater* (20<sup>th</sup> ed.). Washington, DC: Author.
- Baghdadi, M., Ghaffari, E., & Aminzadeh, B. (2016). Removal of carbamazepine from municipal wastewater effluent using optimally synthesized magnetic activated carbon: Adsorption and sedimentation kinetic studies. *Journal of Environmental Chemical Engineering*, 4(3), 3309-3321. doi:10.1016/j.jece.2016.06.034
- Belaid, C., Khadraoui, M., Mseddi, S., Kallel, M., Elleuch, B., & Fauvarque, J. F. (2013). Electrochemical treatment of olive mill wastewater: Treatment extent and effluent phenolic compounds monitoring using

- some uncommon analytical tools. *Journal of Environmental Sciences*, 25(1), 220-230. doi:10.1016/S1001-0742(12)60037-0
- Bensalah, N., Louhichi, B., & Abdel-Wahab, A. (2012). Electrochemical oxidation of succinic acid in aqueous solutions using boron doped diamond anodes. *International Journal of Environmental Science and Technology*, 9, 135-143. doi:10.1007/s13762-011-0007-5
- Bilińska, L., Gmurek, M., & Ledakowicz, S. (2016). Comparison between industrial and simulated textile wastewater treatment by AOPs—Biodegradability, toxicity and cost assessment. *Chemical Engineering Journal*, 306, 550-559. doi:10.1016/j.cej.2016.07.100
- Bolzonella, D., Fatone, F., di Fabio, S., & Cecchi, F. (2010). Application of membrane bioreactor technology for wastewater treatment and reuse in the Mediterranean region: Focusing on removal efficiency of non-conventional pollutants. *Journal of Environmental Management*, 91(12), 2424-2431. doi:10.1016/j.cej.2016.07.100
- Brillas, E., Calpe, J. C., & Casado, J. (2000). Mineralization of 2, 4-d by advanced electro-chemical oxidation processes. *Water Research*, 34, 2253. doi:10.1016/S0043-1354(99)00396-6
- Brillas, E., & Martínez-Huitle, C. A. (2015). Decontamination of wastewaters containing synthetic organic dyes by electrochemical methods. An updated review. *Applied Catalysis B*, 166-167, 603-643.
- Canizares, P., Jimenez, C., Martinez, F., Saez, C., & Rodrigo, M. A. (2007). Study of the electrocoagulation process using aluminium and iron electrodes. *Industrial and Engineering Chemistry*, 46(19), 6189-6195. doi:10.1021/ie070059f
- Cañizares, P., Louhichi, B., Gadri, A. Nasr, B., Paz, R., Rodrigo, M. A., & Saez, C. (2007). Electrochemical treatment of the pollutants generated in an ink-manufacturing. *Journal of Hazardous Materials*, 146(3), 552-557. doi:10.1016/j.jhazmat.2007.04.085
- Canizares, P., Paz, R., Saez, C., & Rodrigo, M. A. (2009). Costs of the electrochemical oxidation of wastewaters: A comparison with ozonation and Fenton oxidation processes. *Journal of Environmental Management*, 90(1), 410-420. doi:10.1016/j.jenvman.2007.10.010
- Devi, P., & Saroha, A. K. (2017). Utilization of sludge based adsorbents for the removal of various pollutants: A review. *Science of The Total Environment*, 578, 16-33. doi:10.1016/j.scitotenv.2016.10.220
- Dinçer, A. R., & Kargı F. (2001). Performance of rotating biological disc system treating saline wastewater. *Process Biochemistry*, 36(8-9), 901-906. doi:10.1016/S0032-9592(00)00287-9
- Dionisi, D., Bruce, S. S., & Barraclough, M. J. (2014). Effect of pH adjustment, solid-liquid separation and chitosan adsorption on pollutants removal from potale wastewaters. *Journal of Environmental Chemical Engineering*, 2(4), 1929-1936. doi:10.1016/j.jece.2014.08.013
- Hai, F. I., Nghiem, L. D., & Modin, O. (2013). 20-Biocatalytic membrane reactors for the removal of recalcitrant and emerging pollutants from wastewater. *Handbook of Membrane Reactors*, 763-807. doi:10.1533/9780857097347.4.763
- Hammadi, B., Bebbba, A. A., & Gherraf, N. (2016). Degradation of organic pollution aerated lagoons in an arid climate: the case the treatment plant Ouargla (Algeria). *Acta Ecologica Sinica*, 36(4), 275-279. doi:10.1016/j.chnaes.2016.05.002
- Hu, J., Aarts, A., Shang, R., Bas, H., & Rietveld, L. (2016). Integrating powdered activated carbon into wastewater tertiary filter for micro-pollutant removal. *Journal of Environmental Management*, 177, 45-52. doi:10.1016/j.jenvman.2016.04.003
- Koprivanac, N., Locke, B. R., Papić, S., LončarićBožić, A., Vujević, D., & Lazarević, Z. (2004). Effect of zeolites in AOPs for dye wastewater treatment. *Studies in Surface Science and Catalysis*, 154(C), 2548-2554. doi:10.1016/S0167-2991(04)80522-6
- Lakshmanan, D., Clifford, D. A., & Samanta, G. (2009). Ferrous and ferric ion generation during iron electrocoagulation. *Environmental Science and Technology*, 43, 3853-3859. doi:10.1021/es8036669
- Liakos, T. I., & Lazaridis, N. K. (2014). Melanoidins removal from simulated and real wastewaters by coagulation and electro-flotation. *Chemical Engineering Journal*, 242, 269-277. doi:10.1016/j.cej.2014.01.003
- Liu, J., & Mattiasson, B. (2002). Microbial BOD sensors for wastewater analysis. *Water Research*, 36(15), 3786-3802. doi:10.1016/S0043-1354(02)00101-X
- Llanos, J., Camarillo, R., Perez, A., Canizares, P., & Rodrigo, M. A. (2011). Costs estimation of an integrated process for the treatment of heavy-metal loaded aqueous effluents. *The Journal of Applied Electrochemistry*, 41(9), 1099-1107. doi:10.1007/s10800-011-0278-z
- Lu, J., Wang, X., Shan, B. Li, X., & Wan, W. (2006). Analysis of chemical compositions contributable to chemical oxygen demand (COD) of oilfield produced water. *Chemosphere*, 62, 322-331.
- Mansouri, K., Elsaid, K., Bedoui, A., Bensalah, N., & Abdel-Wahab, A. (2011). Application of electrochemically dissolved iron in the removal of tannic acid from water. *Chemical Engineering Journal*, 172(2-3), 970-976. doi:10.1016/j.cej.2011.07.009
- Martínez-Huitle, C. A., Rodrigo, M. A., Sires, I., & Scialdone, O. (2015). Single and coupled electrochemical processes and reactors for the abatement of organic water pollutants: A critical review. *Chemical Reviews*, 115, 13362-13407.
- Subba Rao, A. N., & Venkatarangaiah, V. T. (2014). Metal oxide-coated anodes in wastewater treatment. *Environmental Science and Pollution Research*, 21, 3197-3217.
- Sirés, I., Brillas, E., Oturan, M. A., Rodrigo, M. A., & Panizza, M. (2014). Electrochemical advanced oxidation processes: Today and tomorrow. *Environmental Science and Pollution Research*, 21, 8336-8367.



- Panizza, M., & Cerisola, G. (2009). Direct and mediated anodic oxidation of organic pollutants. *Chemical Reviews*, *109*, 6541-6569.
- Martínez-Huitile, C. A., & Ferro, S. (2006). Electrochemical oxidation of organic pollutants for the wastewater treatment: Direct and indirect processes. *Chemical Society Reviews*, *35*, 1324-1340. doi:10.1039/b517632h
- Montalvo, S., Guerrero, L., Rivera, E., Borja, R., Chica, A., & Martín, A. (2010). Kinetic evaluation and performance of pilot-scale fed-batch aerated lagoons treating winery wastewaters. *Bioresource Technology*, *101*(10), 3452-3456. doi:10.1016/j.biortech.2009.12.101
- Moura, A., Tação, M., Henriques, I., Dias, J., Ferreira, P., & Correia, A. (2009). Characterization of bacterial diversity in two aerated lagoons of a wastewater treatment plant using PCR-DGGE analysis. *Microbiological Research*, *164*(5), 560-569. doi:10.1016/j.micres.2007.06.005
- Özcan, A., Şahin, Y., Kopal, A. S., & Oturan, M. A. (2009). Electro-fenton removal of the cationic dye basic blue 3 by using carbon felt cathode. *Environmental Engineering and Management Journal*, *19*(5), 267-275.
- Palma, L. D., & Verdone, N. (2009). The effect of disk rotational speed on oxygen transfer in rotating biological contactors. *Bioresource Technology*, *100*(3), 1467-1470. doi:10.1016/j.biortech.2008.07.058
- Pignatello, J. J., Oliveros, E., & Mackay, A. (2006). Advanced oxidation processes for organic contaminant destruction based on the Fenton reaction and related chemistry. Critical review. *Environmental Science and Technology*, *36*, 1-84. doi:10.1080/10643380500326564
- Pitakpoolsil, W., & Hunsom, M. (2013). Adsorption of pollutants from biodiesel wastewater using chitosan flakes. *Journal of the Taiwan Institute of Chemical Engineers*, *44*(6), 963-971. doi:10.1016/j.jtice.2013.02.009
- Rodríguez, S., Santos, A., & Romero, A. (2011). Effectiveness of AOP's on abatement of emerging pollutants and their oxidation intermediates: Nicotine removal with Fenton's Reagent. *Desalination*, *280*(1-3), 108-113. doi:10.1016/j.desal.2011.06.055
- Rueda-Márquez, J. J., Levchuk, I., Salcedo, I., Acevedo-Merino, A., & Manzano, M. A. (2016). Post-treatment of refinery wastewater effluent using a combination of AOPs (H<sub>2</sub>O<sub>2</sub> photolysis and catalytic wet peroxide oxidation) for possible water reuse. Comparison of low and medium pressure lamp performance. *Water Research*, *91*, 86-96. doi:10.1016/j.watres.2015.12.051
- Soares, P. A., Tânia, F. C. V., Silva, Arcy, A. R., Selene, M. A., Souza, G. U., Boaventura, R. A. R., & Vilar, V. J. P. (2016). Assessment of AOPs as a polishing step in the decolourisation of bio-treated textile wastewater: Technical and economic considerations. *Journal of Photochemistry and Photobiology A: Chemistry*, *317*, 26-38. doi:10.1016/j.jphotochem.2015.10.017
- Wang, L., Liu, J., Zhao, Q., Wei, W., & Sun, Y. (2016). Comparative study of wastewater treatment and nutrient recycle via activated sludge, microalgae and combination systems. *Bioresource Technology*, *211*, 1-5. doi:10.1016/j.biortech.2016.03.048
- Wang, N., Zheng, T., Zhang, G., & Wang, P. (2016). A review on Fenton-like processes for organic wastewater treatment. *Journal of Environmental Chemical Engineering*, *4*(1), 762-787. doi:10.1016/j.jece.2015.12.016
- Wu, X., Yang, Y., Wu, G., Mao, J., & Zhou, T. (2016). Simulation and optimization of a coking wastewater biological treatment process by activated sludge models (ASM). *Journal of Environmental Management*, *165*, 235-242. doi:10.1016/j.jenvman.2015.09.041
- Zhang, Q., Liu, S., Yang, C., Chen, F., & Lu, S. (2014). Bioreactor consisting of pressurized aeration and dissolved air flotation for domestic wastewater treatment. *Separation and Purification Technology*, *138*, 186-190. doi:10.1016/j.seppur.2014.10.024