



## Tertiary treatment of wastewater by coupling anodic oxidation processes using solar energy and the coagulation–flocculation processes

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### ABSTRACT

Tunisia has been compelled to use treated wastewater in irrigation due continuous increase in demand. Since 1999, the Edissa perimeter in Gabes, Tunisia, has been watered with treated wastewater (TWW) from the Gabes sewage treatment plant. However, it is imperative to assess the quality of this water source. The current study aims to accomplish tertiary treatment of TWW of Gabes sewage treatment plant using alternate processes by combining anodic oxidation processes using solar energy with coagulation–flocculation. Treatment under optimal conditions has shown that the combination of two processes was effective in achieving a reduction in pollution with satisfactory results in terms of elimination of the chemical oxygen demand (100%), total organic carbon (100%), biochemical oxygen demand (86%),  $\text{Cl}^-$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , and  $\text{PO}_4^{3-}$  with abatement rates of (100%). The concentration of heavy metals including (Ni, Al, Cu, Pb, Zn, Fe, Cr, Cd) have been reduced from 45% for Fe to 95% for Ni, and complete elimination of germs.

*Keywords:* Wastewater; Irrigation; Coagulation–flocculation; Anodic oxidation

### 1. Introduction

Countries like Tunisia, which belongs to arid and semi-arid regions are facing serious water shortage problems. To enhance the existing situation, decision-makers since 1960 have considered the reuse of treated wastewater (TWW), with the planning of several irrigated perimeters like Sukra-Ariana 1965, Bir Rommana-Nabeul 1984, and Edissa-Gabes 1999 [1–3]. The water quality shows a significant enhancement after undergoing primary and secondary wastewater

treatment, though a significant presence of metallic and organic compounds exists in the treated wastewater. The use of (TWW) can provide more sources like C, N, P, and K which can positively impact the growth of specific crops. Nevertheless, the quality of soil can be affected because of structural degradation (i.e., salinization splash of clays), and the contamination of soil (involving different factors such as mineral, organic, heavy metals, and bacterial pollution). Hence, reusing this source of water will have a long-term impact on natural resources [4–8]. The overall pollutant

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present in water used for irrigation purposes is not the sole parameter, rather it relies on a complex interplay of mechanism and physico-chemical factors that governs their transportation and distribution. The nature of crops, climate, soil composition along with nature has a crucial role in infiltration and fixing, various horizons of pollutants in soils and impacting their transfer to groundwater and plants. In addition, using it for longer duration can increase pollutants in soil. New technologies that generate less pollution can be useful in this field to avoid problems affecting the soils, as the treated water comes from a biological treatment plan. A number of technologies are developed to enhance the quality of treated wastewater, having the objective to remove all organic heavy metals and bacterial content. Such technologies include a number of methods such as adsorption, coagulation–flocculation processes, electro-coagulation processes, electro-flotation, homogeneous process of chemical oxidation ( $\text{H}_2\text{O}_2/\text{Fe}^{2+}$  and  $\text{H}_2\text{O}_2/\text{O}_3$ ), homogeneous/heterogeneous photo-catalytic processes ( $\text{H}_2\text{O}_2/\text{UV}$ ,  $\text{O}_3/\text{UV}$  and  $\text{Fe}^{2+}/\text{H}_2\text{O}_2/\text{UV}$ ;  $\text{TiO}_2/\text{UV}$ ), ultrasonic oxidation, and anodic oxidation and some combinations of these methods [14–30]. The anodic oxidation process and the coagulation–flocculation processes have proven to be effective ways of removing organic, mineral, and bacterial matter. Anodic oxidation is based on the insoluble electrodes that are referred to as high overvoltage of oxygen such as (BDD, pt, Ti/IrO<sub>2</sub>, Ti/RuO<sub>2</sub>, Ti/SnO<sub>2</sub>, etc.) are commonly used to oxidation several organic molecules present in the wastewater through hydroxyl radicals (OH<sup>•</sup>) which are highly powerful oxidizing species that can be used in the environment [31–35]. In addition, anodic oxidation can have a bactericidal effect on bacteria [36]. The coagulation and flocculation processes constitute the basic treatments applied to treat the inert particulate fractions (silts, clays, colloids) or living (planktonic microalgae; micro-invertebrates) and organic macromolecules and certain heavy metals [37]. Abdul Aziz et al. [38] revealed an effective removal efficiency of coagulation and flocculation processes to remove the color from semi-aerobic landfill leachate from one of the landfills in Malaysia through four types of coagulants, namely aluminum (III) sulfate (alum), ferric (III) chloride, ferrous (II) sulfate and ferric (III) sulfate. The main objective of Abdul Aziz's study was to investigate the tertiary treatment of the TWW from the Gabes sewage treatment plant by combining anodic oxidation processes on three different anodes boron-doped diamond (BDD), pt, TiO<sub>2</sub> under solar energy and coagulation–flocculation processes to attain satisfactory results in

terms of elimination of chemical oxygen demand (COD), total organic carbon (TOC), biochemical oxygen demand (BOD<sub>5</sub>), Cl<sup>-</sup>, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup> and PO<sub>4</sub><sup>3-</sup>, heavy metals and germs.

## 2. Materials and methods

### 2.1. Chemicals

All substances were acquired from Panreac and Fluka. A de-ionized source of water was used for aqueous solution which was obtained from the Heal Force Super Series ultra-pure water system.

### 2.2. Experimental set-up

An electrochemical cell was used for performing the electrochemical treatment, using a stirred discontinuous reactor with cathode (iron) and anodes (BDD or pt or TiO<sub>2</sub>) electrodes. These two electrodes had a similar surface area (4 cm<sup>2</sup>), but also had a gap of 1 cm between them. The volume of solution was 0.5 dm<sup>3</sup>, and the temperature was carefully maintained at 25°C. Similarly, during the experiments, magnetic stirring was also maintained. Under galvanostatic conditions, an anodic experiment was conducted and supported using photovoltaic panels (Table 1). Overall, duration was 180 min at 21 for treatment, and during regular intervals, samples were collected.

### 2.3. Jar test

The sedimentation jar test was used to determine the optimum dosage of coagulant (AlOH<sub>3</sub>).

### 2.4. Analytical techniques

The electrical conductivity (EC) and pH of aqueous solutions were measured with the help of a digital thermo pH meter (model BE105, BICASA), which was equipped Metrohm combined glass electrode. For determining COD, a PhotoLab VWR S12 analyzer based on the dichromate oxidation method was considered. The carbon concentration was determined using a Behr TOC TF 50 analyzer. A turbid meter (Hanna Instruments HI83414 model) was used for measuring turbidity, an oximeter (Hanna Instruments HI5421 model) was used for measuring the levels of dissolved oxygen, and the Indophenol blue spectrophotometric method was taken in consideration to determine NH<sub>4</sub><sup>+</sup> concentrations. Similarly, PO<sub>4</sub><sup>-</sup> and NO<sub>3</sub><sup>-</sup> were quantified using ion chromatography (Metrohm 850 professional IC model). Lastly, the metal contents including (Ni, Cu, Al, Cu, Pb, Fe, Cd, and Cr) in water samples were evaluated with the help of AAS (Atomic Absorption Spectrophotometry) using Metrohm polarograph (7460 VA Trace Analyzers) and VARIO6-type graphite furnace.

### 2.5. Microbiologic analyses

Baird Parker medium was applied for *Staphylococcus aureus* as a culture medium. It involves growth promoting and a rich nutrient base glycine and sodium pyruvate. In case of total coliforms (TC) and *Escherichia coli* the culture

Table 1  
Characteristics of the photovoltaic panels used

Matter	Poly-crystalline silicon
Maximum power ( $P_m$ ), W	2
Maximum power voltages ( $V_{pm}$ ), V	8
Maximum power current ( $I_{pm}$ ), A	0.25
Short circuit current ( $I_{sc}$ ), A	0.263
Open circuit voltage ( $V_{oc}$ ), V	9.2
Maximum system voltage, V	500
Dimensions, mm	170 × 130 × 4

medium used was triphenyltetrazolium chloride (TTC) tergitol, primarily used in water colorimetry through the filtration technique.

### 3. Results and discussion

#### 3.1. Treated wastewater quality

To have a clear concept of the physico-chemical and microbial quality of the treated wastewater used in the irrigated areas, a statistical study was carried out over 24 months. Table 1 provides details on the quality of wastewater from the Gabes wastewater treatment plant. A statistical analysis of the data shows a stable composition of the treated wastewater over the studied period. The findings showed that the average EC is  $4.5 \text{ mmhos}\cdot\text{cm}^{-1}$  and the pH level remains slightly alkaline with an average value of 7.8. In addition, the content of suspended soil referred to as material in suspension (MS), measures at  $2.7 \text{ mg}\cdot\text{L}^{-1}$ . Additionally, the concentrations of both BOD and COD are recorded, average between  $87.9$  and  $18.5 \text{ mg}\cdot\text{L}^{-1}$ , respectively. The  $\text{BOD}_5/\text{COD}$  ratio was equal to 0.21. All these parameters were pored very similarly compared to Tunisian Standards (TS). The mean of  $\text{Cl}^-$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , and  $\text{PO}_4^{3-}$  content was lower than the Tunisian Standards. Besides, the following heavy metals (Cr, Fe, Zn, Pb, Cu, Al and Ni) were lower than the Tunisian Standard average. However, Cd is very high compared to Tunisian Standards.

#### 3.2. Treatment of wastewater solutions by anodic oxidation

##### 3.2.1. COD and TOC degradation

Figs. 1 and 2 show the effect of the percent removal of COD and TOC from three different electrodes (BDD, pt,  $\text{TiO}_2$ ) with a particular electric charge ( $Q$ ) during galvanostatic electrolysis of treated wastewater used in irrigated areas.

$$Q(\text{Ahdm}^{-3}) = \frac{I \cdot t}{3600 \cdot V} \quad (1)$$

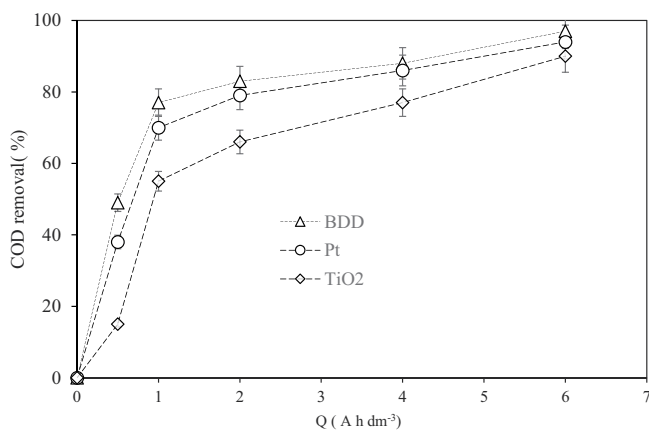


Fig. 1. COD efficiency with specific electric charge of the effluent treated by anodic oxidation;  $\text{COD}_0 = 89 \text{ mg}\cdot\text{L}^{-1}$ ; electrodes (BDD, pt,  $\text{TiO}_2$ );  $I = 2\text{A}$ ;  $T = 25^\circ\text{C}$ .

where  $I$  is current intensity (in A),  $t$  is time (in s), and  $V$  is reactor volume (in  $\text{dm}^3$ ). The reduction in COD and TOC was very quick at the beginning of treatment up to  $2 \text{ A}\cdot\text{h}\cdot\text{L}^{-1}$ . BDD electrode showed a slight increase in organic matter degradation followed by the pt electrode and then the  $\text{TiO}_2$  electrode. This is because of the massive production of  $\text{OH}^\bullet$  radicals by the BDD anode compared to the other electrodes. After, they become lower as the specific electric charge ( $Q$ ) increases. It should be noted that towards the end of the electrochemical treatment, the COD and the TOC were sufficiently destroyed. Certainly, when subjected to specific electric charges of less than  $6 \text{ A}\cdot\text{h}\cdot\text{dm}^{-3}$ , the COD removal efficiency shows an increase to 90%, 94%, and 97% (Fig. 1), and the TOC removal efficiency show an increase to 82%, 90%, and 94% (Fig. 2) with the use of  $\text{TiO}_2$ , pt, and BDD electrodes, respectively. In fact, the organic matter found in the wastewater is organic compounds with a benzene cycle, saturated and unsaturated linear carbon chains. The presence of  $\text{OH}^\bullet$  radicals leads to the breaking of the benzene ring and the substitution of the double or triple bonds by two or more hydroxyl groups. Furthermore, the constant reduction in TOC through the organic matter treatment on the anodes initiates the production of  $\text{CO}_2$  right from the beginning of electrolysis. Nevertheless, minimal quantities of refractory carbon were found towards the concluding part of the treatment. This implies that the formation of different intermediates, indicating  $\text{CO}_2$  is not entirely the result of compound oxidation, rather it suggests the presence of simpler structures of the company ranging from a minor oxidation state of carbon ( $\text{CH}_4$ ) and +4 for the major oxidation state ( $\text{CO}_2$ ) [35].

##### 3.2.2. pH evolution

Fig. 3 reveals the pH changes in the solution over the course of the electrolysis process. At the start of the treatment, there was a slight reduction in the pH, however, subsequently, it resumed to its original position. This can be explained by the formation of shorter-chain compounds and the formation of  $\text{CO}_2$ . The rapid recovery of pH can be explained by the buffer effect of water and the decrease in

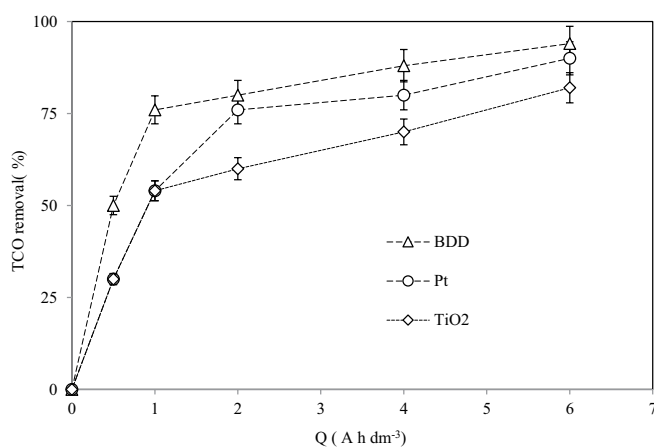


Fig. 2. TOC efficiency with specific electric charge of the effluent treated by anodic oxidation;  $\text{COD}_0 = 89 \text{ mg}\cdot\text{L}^{-1}$ ; electrodes (BDD, pt,  $\text{TiO}_2$ );  $I = 2\text{A}$ ;  $T = 25^\circ\text{C}$ .

the concentration of compounds formed due to the degradation contained on the electrodes.

This phenomenon can be elucidated by the reaction occurring at the anode, which encourages the production of OH• radicals. In addition, the radicals promote short-chained carboxylic acid production and attack organic matter, which acidifies the medium. However, the subsequent rise in pH is attributed to the degradation of these acids.

### 3.2.3. BOD<sub>5</sub> degradation and bacteria removal

The BOD<sub>5</sub> decreased under 10 mg·dm<sup>-3</sup> following anodic oxidation by the three electrodes (Fig. 4). This can be elucidated by bacteria destruction and the degradation of organic matter due to the effect of OH• radicals. In addition, the bacteria are at risk due to the flow of current passing through wastewater. In Table 2, a summary of microbiological analyses of bacteria (*Staphylococcus aureus*, *Escherichia coli* and TC) is provided, both after and before treatment by anodic oxidation with BDD, pt, and TiO<sub>2</sub> electrodes. The total absence of microorganisms is noted at the end of treatment. This is due to the direct electrolysis on bacteria and to the OH• generation, which extinguishes and kills microorganisms. A study by Gaied et al. [36] indicated

that the bacteria count decreased for (GMT, *Escherichia coli*, Staph and Salmonella) after treatment by ECiron, EFiron, and AEOPt processes. Other studies have shown that electrolysis of concentrated suspensions of *Escherichia coli*, on active agglomerated porous carbon electrodes, leads to the deactivation of bacteria. On either side of the bacterial membranes, there is an electric field which, considering the small thickness of these membranes is of the order of a few hundred volts per centimeter. A bacterial effect is observed when an electric field greater than bacterial membrane level is subjected to contaminated solution.

### 3.2.4. Concentration of Cl<sup>-</sup>, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, and PO<sub>4</sub><sup>3-</sup> evolution during the galvanostatic electrolysis

Fig. 4A–C show the evolution of the concentration of Cl<sup>-</sup>, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, and PO<sub>4</sub><sup>3-</sup> during the galvanostatic electrolysis on the electrodes of BDD, pt, and TiO<sub>2</sub>. After consumption of about 2 A·h<sup>-1</sup>·dm<sup>-3</sup>, it noticed a rapid drop in concentration for the elements Cl<sup>-</sup>, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup> in all cases. This can be explained by the mechanism of oxidation on the anode

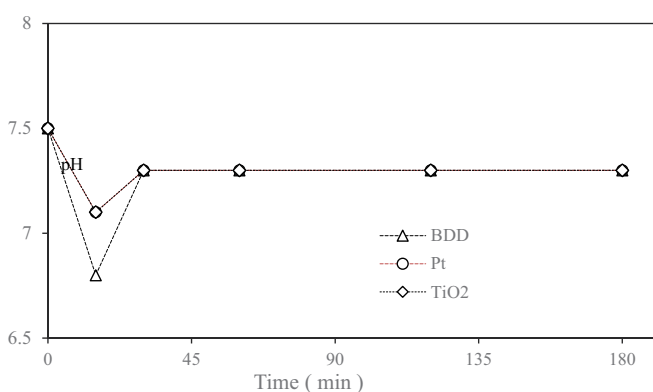


Fig. 3. pH evolution during the electrolysis time ( $I = 2A$ ,  $T = 25^{\circ}C$ ).

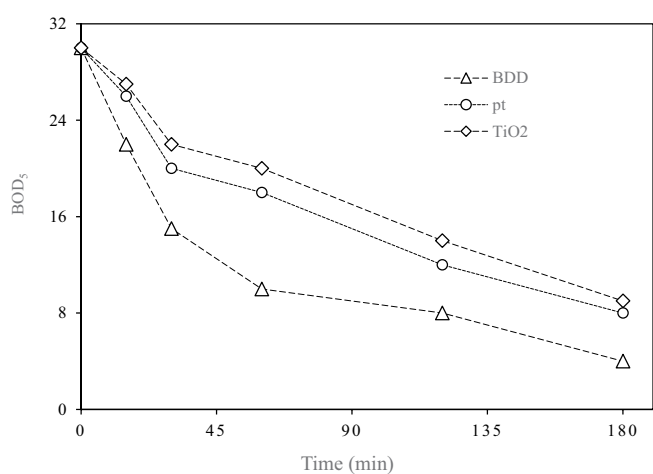


Fig. 4. BOD<sub>5</sub> removal efficiency by anodic oxidation during the electrolysis time ( $I = 2A$ ,  $T = 25^{\circ}C$ ).

Table 2

Mean values of physico-chemical, microbial, and heavy metals contents of the treated wastewater

Parameters	TWW	Standard deviations	TS
EC (mS·cm <sup>-1</sup> )	4.50	0.123	7.00
pH	7.80	0.098	6.50–8.50
MS (mg·L <sup>-1</sup> )	2.70	0.90	30.00
COD (mg·L <sup>-1</sup> )	87.90	15.00	90.00
BOD (mg·L <sup>-1</sup> )	18.50	4.00	30.00
BOD <sub>5</sub> /COD	0.22	–	–
Dissolved oxygen (mg·L <sup>-1</sup> )	0.43	–	–
Cl <sup>-</sup> (mg·L <sup>-1</sup> )	46.00	6.00	2,000
NH <sub>4</sub> <sup>+</sup> (mg·L <sup>-1</sup> )	34.00	3.00	50.00
NO <sub>3</sub> <sup>-</sup> (mg·L <sup>-1</sup> )	6.50	1.22	0.500
PO <sub>4</sub> <sup>3-</sup> (mg·L <sup>-1</sup> )	1	0.56	2.00
Heavy metals			
Cd (μg·L <sup>-1</sup> )	30.00	0.533	10.00
Cr (μg·L <sup>-1</sup> )	10.00	0.410	100.00
Fe (μg·L <sup>-1</sup> )	1,300.00	7.357	5,000.00
Zn (μg·L <sup>-1</sup> )	180.00	1.567	5,000.00
Pb (μg·L <sup>-1</sup> )	2,510.00	9.777	1,000.00
Al (μg·L <sup>-1</sup> )	2,250.00	5.667	5,000.00
Cu (μg·L <sup>-1</sup> )	90.00	1.666	500.00
Ni (μg·L <sup>-1</sup> )	80.00	0.567	200.00
Microbial			
TC (colonies/mL)	7,300	–	–
<i>Escherichia coli</i> (colonies/mL)	1,500	–	2,000
SF (colonies/mL)	1,100	–	1,500

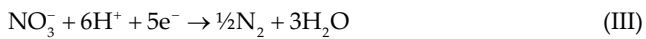
TWW: treated wastewater;  
TS: Tunisian Standards.

and the reduction (R) on the cathode, which can produce  $\text{Cl}_2$  and  $\text{N}_2$ .

3.2.4.1. Anode



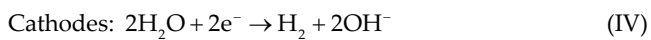
3.2.4.2. Cathode



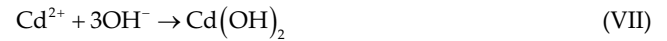
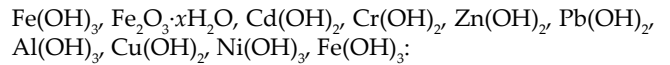
On the other hand, for  $\text{PO}_4^{3-}$  concentration is maintained and it has kept the same level of concentration throughout the slow process and the resistance of this ion to oxidize or to reduce.

3.2.5. Concentration of heavy metals evolution during the galvanostatic electrolysis

Fig. 6 shows a marginal reduction in the concentration of various heavy metals within the solution over the course of electrolysis at a constant current intensity (I) of 2A. This decrease is probably because of the transformation of metal ions into hydroxyl formed by combining  $\text{OH}^-$  produced at the cathode.



The resulting metal ions are combined with the  $\text{OH}^-$  formed at the cathode to produce insoluble forms  $\text{Fe}(\text{OH})_2$ ,



However, no gelatinous metallic coagulant was detected in the solution during treatment. This can be explained by the rapid recovery of new forms in their initial ionic form under the effect of anodic oxidation.

3.3. Treatment of wastewater solutions by the coagulation–flocculation processes

3.3.1. Optimum dose of coagulant

Fig. 7A and B show the variations in iron and aluminum concentrations in response to changing coagulant ( $\text{Al}(\text{OH})_3$ ) dosages. The minimum concentration of both elements was attained at a coagulant dose of 40  $\text{mg}\cdot\text{L}^{-1}$ . Especially, the iron concentration was reduced from 1,100 to 500  $\text{mg}$ , while the aluminum concentration was reduced

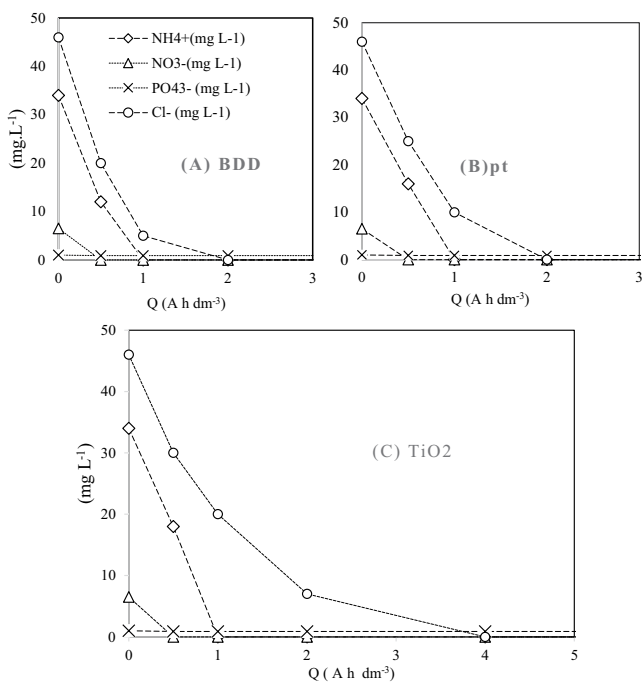


Fig. 5.  $\text{Cl}^-$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , and  $\text{PO}_4^{3-}$  evolution with specific electric charge during the galvanostatic electrolysis.

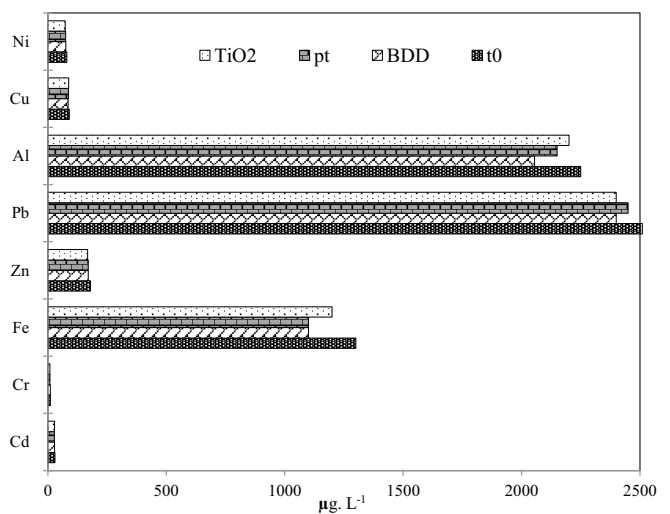


Fig. 6. Heavy-metals evolution before and after galvanostatic electrolysis.

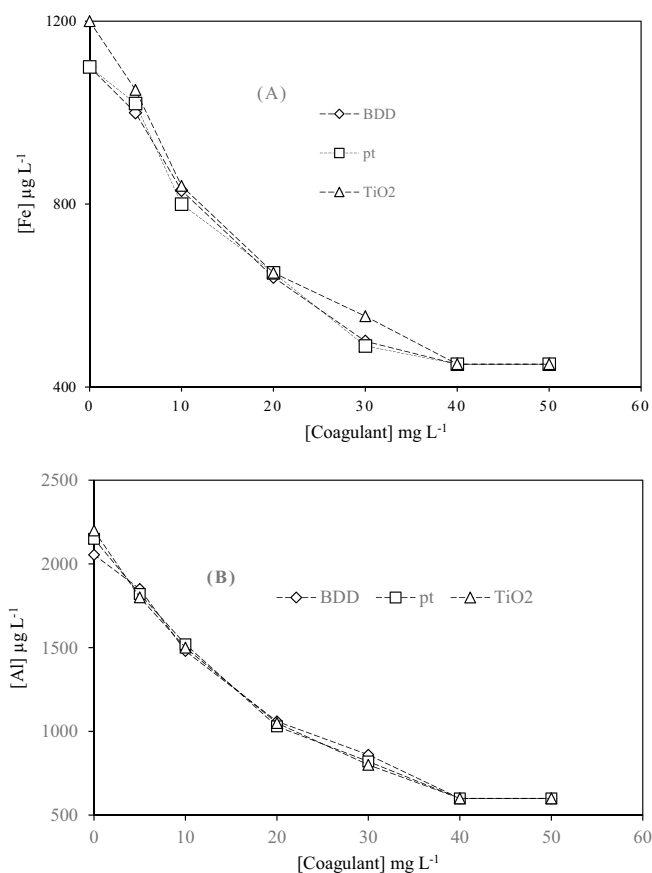


Fig. 7. Variation of the iron concentration and the aluminum with the dose of coagulant (AlOH<sub>3</sub>).

Table 3  
Microbiologic analyze before and after anodic oxidation

Bacteria	Time	<i>t</i> <sub>0</sub>	<i>t</i> <sub>180</sub> /BDD	<i>t</i> <sub>180</sub> /pt	<i>t</i> <sub>180</sub> /TiO <sub>2</sub>
	TC (colonies/mL)		7,300	<1.0	<1.0
<i>Escherichia coli</i> (colonies/mL)		1,500	<1.0	<1.0	<1.0
SF (colonies/mL)		1,100	<1.0	<1.0	<1.0

from 2,200 to 1,000 mg. In addition, we assessed the concentration of different heavy metals at a coagulant dose of 40 mg. The results in Table 3 reveal significant reductions in the concentrations of these metal ions. The percentage of heavy metal removal exhibited considerable variation: 45% for Fe, 67%–78% for Cd, 77%–88% for Cr, 64%–70% for Zn, 75% for Pb, 75%–81% for Al, 88%–90% for Cu, and an impressive 95% for Ni.

### 3.4. Water quality after coupling anodic oxidation processes and the coagulation–flocculation processes

Table 4 illustrates the physico-chemical and microbial quality of the water after combining anodic oxidation processes with coagulation–flocculation processes. There is a significant improvement in water quality at all levels of

Table 4  
Heavy metal removal efficiency

Heavy metal	Heavy metal removal efficiency (%) / BDD	Heavy metal removal efficiency (%) / pt	Heavy metal removal efficiency (%) / TiO <sub>2</sub>
Cd	78	67	78
Cr	80	77	77
Fe	45	45	45
Zn	70	64	70
Pb	75	75	75
Al	75	76	81
Cu	90	88	88
Ni	92	92	95

Table 5  
Mean values of physico-chemical, microbial, and heavy metals contents of the water

Parameters	BDD	pt	TiO <sub>2</sub>
EC (mS·cm <sup>-1</sup> )	3.00	3.00	3.00
pH	7.30	7.30	7.30
MS (mg·L <sup>-1</sup> )	0.00	0.00	0.00
COD (mg·L <sup>-1</sup> )	0.00	0.00	0.00
BOD (mg·L <sup>-1</sup> )	4.00	4.00	4.00
Cl <sup>-</sup> (mg·L <sup>-1</sup> )	0.00	0.00	0.00
NH <sub>4</sub> <sup>+</sup> (mg·L <sup>-1</sup> )	0.00	0.00	0.00
NO <sub>3</sub> <sup>-</sup> (mg·L <sup>-1</sup> )	0.00	0.00	0.00
PO <sub>4</sub> <sup>3-</sup> (mg·L <sup>-1</sup> )	0.00	0.00	0.00
Heavy metals			
Cd (µg·L <sup>-1</sup> )	6.00	8.00	6.00
Cr (µg·L <sup>-1</sup> )	2.00	2.00	2.00
Fe (µg·L <sup>-1</sup> )	600.00	600.00	600.00
Zn (µg·L <sup>-1</sup> )	50.00	55.00	50.00
Pb (µg·L <sup>-1</sup> )	600.00	600.00	600.00
Al (µg·L <sup>-1</sup> )	500.00	500.00	500.00
Cu (µg·L <sup>-1</sup> )	8.00	10.00	10.00
Ni (µg·L <sup>-1</sup> )	6.00	6.00	6.00
Microbial			
TC (colonies/mL)	<1.0	<1.0	<1.0
<i>Escherichia coli</i> (colonies/mL)	<1.0	<1.0	<1.0
SF (colonies/mL)	<1.0	<1.0	<1.0

organic load, microbial, and heavy metals. It was possible to remove all organic and microbial matter. However, the BOD<sub>5</sub> has a reduction efficiency of close to 86% and heavy metals have been significantly reduced.

## 4. Conclusion

This study has investigated the tertiary treatment on the TWW Gabes wastewater treatment plant for irrigation purposes. It involves the combination of anodic oxidation

processes using coagulation and energy flocculation processes, yielding in the following outcomes;

- COD and TOC removal efficiency increased in the treated wastewater up to 100%.
- BOD<sub>5</sub> removal efficiency reached 86%.
- Cl<sup>-</sup>, NH<sub>4</sub><sup>+</sup>, and NO<sub>3</sub><sup>-</sup> were eliminated by anodic oxidation however PO<sub>4</sub><sup>3-</sup> was eliminated by the coagulation–flocculation processes.
- Most of the heavy material was removed by the coagulation–flocculation processes and varies from 45% to 95%.
- Total absence of bacteria (TC, *Escherichia coli*, and *Staphylococcus aureus*) was noted at the end of treatment due to the direct electrolysis on bacteria and to the OH<sup>•</sup> generation.

The present tertiary treatment has shown a great capacity to reduce organic, mineral, and bacterial loads. This water could be then used for irrigation with less risk to soil and humans. A technical and economic study is necessary to determine the choice of electrode at the industrial level.

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