



## Phenol Deterioration in Refinery Wastewater through Advanced Electrochemical Oxidation Reactions Using Different Carbon Fiber and Graphite Electrodes Configurations

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### Abstract

Phenolic wastewater was treated electrochemically by various cells consist different arrangements of carbon fiber and graphite electrodes by direct, indirect, and Fenton electrochemical oxidation processes. The oxidation process carried at 35 °C for 180 minutes and at 8 mA/cm<sup>2</sup> current density. One gram of NaCl per liter of electrolyte and 0.4 mM of ferrous ion concentration were used as a catalyst in the indirect and Fenton electrochemical oxidation process, respectively. The results indicated that the effectiveness of graphite-graphite cells in removing phenol via direct and indirect electrochemical removal was the highest of the other cells. In contrast, the cell of a graphite anode and a carbon fiber cathode was the most efficient in removing phenol using the electro-Fenton process, and 55.59% of phenol was removed after 180 minutes. This significant increase in the removal via the electro-Fenton process using carbon fiber as a cathode was due to the large surface area of the cathode, which improves the hydrogen peroxide production rate. The results of the kinetics study of phenol removal by different oxidation processes and by using other cells in the arrangement of the electrodes indicated that the phenol removal reactions followed the first-order kinetics. The extensive study of the reaction rate constant values showed that the electro-Fenton process in a cell of graphite (as an anode) and carbon fiber (as a cathode) cell was faster than the other electro-oxidation processes and cells. Specific power consumption (SPC) was also investigated and showed that the better arrangement is using graphite as an anode that gives the lowest SPC ( 57.1 kWh/kg phenol).

Keywords: Phenol removal; refinery wastewater; electrochemical; oxidation; carbon fiber; graphite

### 1. Introduction

Phenols are a large group of pollutants in industrial wastewater [1] and are considered a series of the most severe and persistent organic pollutants for biodegradable [2]. Correspondingly, phenolic pollutants have low degradability by conventional treatment and toxic effects on human and aquatic life. In some cases, phenol becomes fatal when ingested or inhaled and its vapors or adsorbed by skin [3], [4]. Phenol in high concentration can cause severe burns and harmful effects on the lung, liver, heart, and central nervous system. Phenol and its vapors are corrosive to the eyes, the skin, and the respiratory tract. Repeated or prolonged skin contact with phenol may cause dermatitis or second and third-degree burns due to phenol caustic and defatting properties [5].

Phenolic compounds are used widely in several industrial activities such as resin and polymer

manufacturing, pesticides, paper pulp, petroleum and petrochemical, coal-conversion, drugs, and other synthetic products [6]–[9]. Petroleum refinery plants consume about 65 to 90 gallons (246 to 341 L) of water per barrel of crude [10], and nearly 0.4–1.6 times the volume of the processed crude oil is discharged as petroleum refinery wastewater [11]. The removal of phenol and phenolic wastewater is conducted by the methods of extraction [6], [12], absorption [13], ionic exchange [14], membrane separation [15], adsorption [16], [17], oxidation with the use of chemical reagents [18], [19], biological and enzymatic treatments [20], ozone oxidation [21], ultrasonic degradation [22], [23], electrochemical oxidation [9], [24], electro-Fenton oxidation [25]–[27].

Electro-oxidation processes for treating the organic matters in wastewater have significant advantages, the most important of which are ease of operation, high

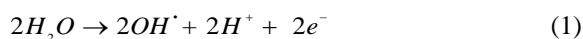
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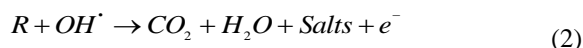
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efficiency, and compatibility with the environment [28]–[30]. Therefore, advanced electrochemical oxidation processes (EAOPs) have increased global attention to treat persistent organic materials such as phenol and its derivatives [31]. In addition, these processes are cost-effective [9], [32] as the oxidizer responsible for the removal process is obtained on-site [33].

Electrochemical oxidation techniques include direct oxidation, indirect oxidation, and electro-Fenton oxidation. During the direct oxidation process, the pollutants diffuse from the bulk solution to the anode surface, where the oxidation occurs [34]. The organic contaminants are oxidized on the anode surface by the adsorbed hydroxyl radicals electrogenerated by water as Eq. (1) [35].



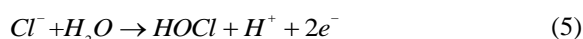
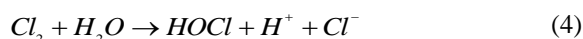
Organic matters are converted into carbon dioxide, water, and other inorganic substances, as presented by Eq. (2) [36].



In indirect oxidation, efficient oxidizing agents such as chlorine are generated to destroy oxidizable pollutants [37]. Chloride salts of potassium or sodium in indirect electro-oxidation are added to the wastewater to promote conductivity and generation of hypochlorite ions. The reaction of anodic oxidation of chloride ions to form chlorine is given in Eq. (3) [38], [39]:



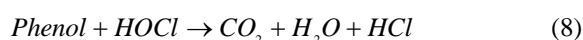
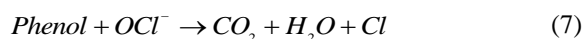
According to Eq. (4) and (5), the released chlorine forms hypochlorous acid at bulk solution reactions.



And further dissociates gives hypochlorite ion as shown in Eq. (6).

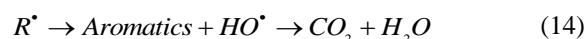
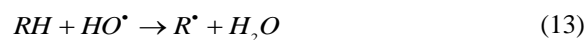
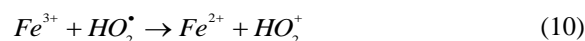


Organic matters, such as phenol, are continually destroyed by reacting with hypochlorous acid and hypochlorite ion. The reactions are converted organic into intermediates, then to carbon dioxide and water, according to Eq. (7) and (8).



Electro-Fenton oxidation is consisting of either adding  $Fe^{2+}$  or reduction of  $Fe^{3+}$  to  $Fe^{2+}$  electrochemically with the simultaneous production of

$H_2O_2$  upon the reduction of  $O_2$  on the electrodes. Compared with traditional Fenton-reagent oxidation, electro-Fenton can avoid the high cost of  $H_2O_2$ , maintain an almost constant concentration of  $H_2O_2$  and regenerate  $Fe^{2+}$  more effectively. Electro-Fenton can oxidize organic compounds quickly and economically [26], [40]. The following reactions Eq. (9) through Eq. (14), occur in the electro-Fenton process [26].



The performance of the electrolytic system depends on several parameters, among which the electrode material is the most important [26]. The electrodes play major roles in organics degradation, and different materials of the electrode lead to different removal efficiency even under the same operating condition [41]. Several electrode materials, such as graphite [26], [32], [42] Nobel metals [40], [43], and transition metal [44], [45] are used in the oxidation of organic pollutants [38].

Stable electrodes used in the electrochemical reactions enable the generation of weakly adsorbed hydroxyl radicals that can oxidize organic pollutants. Among the electrodes that provide excellent results in the electro-oxidation of organic molecules are those with a high overpotential oxygen evolution reaction. Boron-doped diamond (BDD) [25], [46], [47],  $SnO_2$ -Sb [48], [49],  $PbO_2$  [2], [50], and carbonaceous anodes (e.g., glassy carbon [51], carbon felt [52], granular carbon [53]) are suitable for this purpose. However, although the BDD is chemically stable and shows excellent results in the degradation of organic compounds, it is expensive, while the  $SnO_2$ -Sb electrode becomes deactivated after a period of use [54].

Carbon electrodes such as carbon fiber [55], activated carbon fiber [3], graphite [56], activated carbon [31], [57], carbon nanotube [29], carbon black [58] are used extensively in the electrochemical treatment of wastewater. Carbonaceous material has a high specific surface area, large pore volume, allows the adsorption of phenol on its surface, has good conductivity, also acts as an electron transfer catalyst

[31], [55], [57].

This work aims to test the ability of graphite and unmodified carbon fiber to remove phenol from an aqueous solution with concentration simulated to the organic matter present in the untreated wastewater from the Al-Dora refinery units in Baghdad (Iraq) by advanced electrochemical oxidation processes (direct, indirect, and Fenton). The research will choose the best cell from several electrodes arrangements and choose the most effective oxidation process regarding removal, energetic parameters, and kinetics.

### Experimental work

A dilute phenol solution with a concentration of 150 ppm was prepared to simulate the total amount of organic matter in the untreated wastewater of Al-Dora Refinery in Baghdad. The prepared dilute phenol solution was oxidized using direct, indirect, and electro-Fenton oxidation processes in a 2-liter batch reactor that contained two electrodes separated by a distance of 3 cm. The oxidation processes were carried out using four cells containing graphite (G) imported from Graphite India Limited (GIL) and carbon fiber (CF) purchased from OEM with 0.2 mm thickness and 3k yarn size. The electrodes (130 mm length and 50 mm width) were used as anode and cathode, in the following order: The first cell was used carbon fiber electrodes (CF-CF) as anode and cathode electrodes. As for the second cell, graphite electrodes (G-G) were used as anode and cathode electrodes. In comparison, one of the electrodes was replaced in the other cells to form the third cell containing carbon fiber as the anode and graphite as the cathode electrode (CF-G). The fourth cell used different electrodes in reverse order (G-CF).

Direct, indirect, and electro-Fenton oxidation reactions were utilized at 35 °C, and the mixing of 200 rpm was achieved by using an electrical gearbox stirrer (Heidolph). Sodium sulfate (Na<sub>2</sub>SO<sub>4</sub>) was used as a supporting electrolyte solution in direct and electro-Fenton processes with a concentration of 0.5 and 0.05 M, respectively, and 1 g/l of sodium chloride was used in indirect oxidation. In electro-Fenton oxidation, 0.4mM of FeSO<sub>4</sub>·7H<sub>2</sub>O was added. The initial pH of the simulated solutions (pH=3) was adjusted using 0.1M sulfuric acid (H<sub>2</sub>SO<sub>4</sub>). Finally, samples were collected during the electro-oxidation process at different times up to 180 minutes, and the phenol concentration was measured by using an ultra-violet

visible spectrophotometer (Thermo Genesys 10 UV Electron Corporation Madison W153711). The phenol removal was calculated by Eq. (15). Table 1 states the chemicals used, their sources, states and purity.

$$\text{Phenol removal, \%} = \frac{C_{phe_o} - C_{phe_i}}{C_{phe_o}} \times 100\% \quad (15)$$

Table 1 : Chemicals names, formula, sources, states and purity

Chemicals (formula)	Source	State (purity, %)
Phenol(C <sub>6</sub> H <sub>5</sub> O <sub>6</sub> )	THOMAS BAKER	Colorless crystal (99)
Sodium sulfate (NaSO <sub>4</sub> )	SDFCI	White crystal
Sulfuric acid (H <sub>2</sub> SO <sub>4</sub> )	CHEM-LAB	Liquid (96)
Sodium chloride (NaCl)	HIMEDIA	White crystal (99)
Ferrous sulfate (FeSO <sub>4</sub> ·7H <sub>2</sub> O)	HIMEDIA	Blue-green crystal (99)

### Results and discussion

#### *Effect of electrodes arrangement and techniques on phenol removal*

The oxidation process was carried out at 8 mA/cm<sup>2</sup> current density, and 35 °C, and the results of removal were recorded up to 180 minutes and plotted in Figures 1, 2, and 3 for the four cell configurations and the three electro-oxidation techniques (direct, indirect and Fenton).

From Figures 1 to 3, it is noted that the removal of phenol was higher in cells that use G as an anode, and these removals increase remarkably during the first 30 minutes and continue to increase until it reaches the highest values for each electro-oxidation technique after 180 minutes. The phenol removals of direct oxidation were 30.71% and 30.52% after 180 minutes when using G-G and G-CF cells (as shown in Figure 1), while the removals reached only 3.72% and 5.35% when CF-G and CF-CF electrodes were used. Figure 2 shows that the phenol removals by the indirect electro-oxidation were 44.17%, 43.32%, 6.21%, and 6.99% after 180 minutes when using G-G, G-CF, CF-G, and CF-CF. Similarly, Figure 3 shows the phenol removals after 180 minutes by electro-Fenton oxidation process were 50.74%, 55.59%, 8.11%, and 9.16%. Our results are in agreement with those reported previously [26], [40]. Abbas and Abbas [26] used graphite electrodes after depositing MnO<sub>2</sub> on their surface as an anode in the electro-Fenton oxidation process and the chemical oxygen demand was reduced 57.07% after 6 hours of treatment.

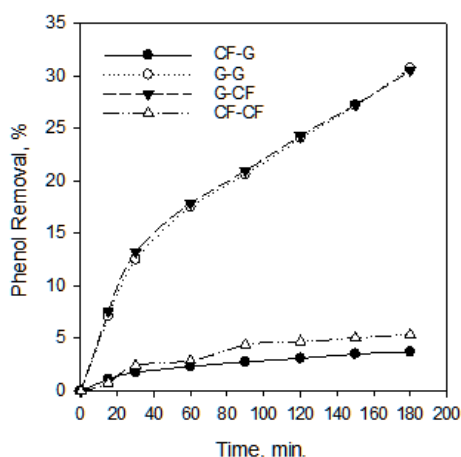


Fig. 1. Removal of phenol via direct oxidation process using the various electrodes' arrangement.

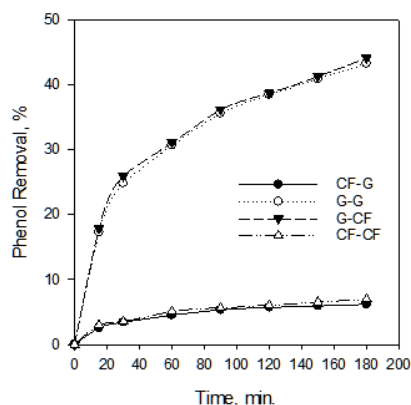


Fig. 2. Removal of phenol via indirect oxidation using the various electrodes' arrangement.

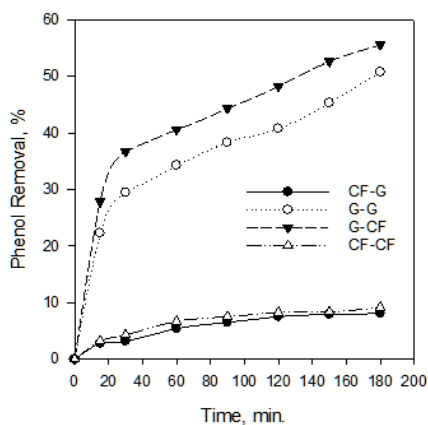


Fig. 3. Removal of phenol via electro-Fenton oxidation process using the various electrodes' arrangement.

The results of the three electrochemical oxidation processes used indicate that G-G electrodes and G-CF electrodes are the most efficient in removing phenol from refinery wastewater. In addition, the use of CF as a cathode has slightly raised the phenol removal, while the cells in which CF was used as the anode did not

give high efficiency in removing phenol.

The CF cathode was not an effective phenol oxidizer in the direct and indirect oxidation processes, even though it generated more hydrogen peroxide. This is because there was no catalyst to transform the hydrogen peroxide into hydroxyl radicals [59].

In the electro-Fenton process, it was observed that the values of phenol removal using a G-CF cell are higher than that of a G-G cell. This improvement in phenol removal was due to the enrichment of the hydrogen peroxide formation, which takes place on the large surface area of the CF cathode. This high surface area causes a remarkable enhancement in the cathodic reduction step for dissolved oxygen or air, according to (Eq. 16) [60], on which the hydroxyl radical production equation depends on interaction with the  $Fe^{2+}$  catalyst (Eq. (9)). Furthermore, CF is a three-dimensional electrode that has a high surface area to volume ratio that is conducive to mass transfer and diffusion of dissolved oxygen with good conductivity and can accumulate a large amount of hydrogen peroxide and also plays a role in the regeneration of ferrous ions [61], [62].



In all arrangements, the direct oxidation has the lowest removal efficiency, and the Fenton process has the highest. Electrode fouling caused by electro-oxidation of phenolic compounds and formation of a passivating film on the electrode surface impedes the practical applications of electrochemical oxidation, especially in the direct oxidation process [63]. The increase in phenol concentration will cause less direct oxidation due to the formation of insoluble compounds or polymers [64], [65], and that will cause a decrease in the surface area of the electrode [66]. Indirect oxidation gives a high performance toward oxidation of the phenolic compounds because of the using sodium chloride as a supporting electrolyte. The pollutants are removed in the bulk solution by the oxidation reaction of the produced oxidant, and that also decreases the energy consumption [66]. The electro-Fenton oxidation process gives the highest performance between the three methods because the degradation of organic compounds is mediated by the strong  $\bullet OH$  radicals produced by catalytic decomposition of electrogenerated hydrogen peroxide ( $H_2O_2$ ), which it is the strongest oxidation radical [26].

### Kinetics of phenol deterioration

The deterioration kinetics of 150 ppm of phenol in wastewater at 8 mA/cm<sup>2</sup> current density and 35 °C and 0.4 mM of ferrous ion concentration as a catalyst in electro-Fenton oxidation was investigated at different oxidation techniques. The oxidation reaction rate constant calculated for the four cells configurations were determined considering the first-order rate expression [7], [27], [67], [68] as in Eq. (17), and the rate constants for all cells and electro-oxidation techniques were summarized in Table 2.

$$\text{rate} = \frac{-d[\text{Cphe}_t]}{dt} = k \cdot [\text{Cphe}_t]^1 \quad (17)$$

The calculated values of the correlation coefficient ( $R^2$ ) showed relatively high values for all cells and electro-oxidation techniques (ranged between 0.9040 and 0.9662), which strongly support the hypothesis that the oxidation reaction follows the first degree.

The reaction constant values indicated that the direct electrochemical oxidation reaction was the slowest technique, even using cells G-CF or G-G. Although, the results of the indirect electrochemical oxidation technique were significantly faster than the indirect oxidation, especially by using cells with anodes of G. In addition, the Fenton process was the fastest of all techniques with the highest reaction rate constants 0.044 and 0.0054 min<sup>-1</sup> for the G-G and G-CF cells, respectively.

### Cells SPC

SPC of cells with their different electrode arrangements and for all types of oxidation processes (direct, indirect, and electro-Fenton method) was calculated at constant applied current per unit mass phenol removed by using Eq. (18) [44], and the results

were illustrated in the Figures 4, 5 and 6.

$$\text{SPC} = \frac{VIt}{V_s \Delta \text{Conc.}} \quad (18)$$

Where V is the cell potential in (volt), I is the applied current in (A), t time of electrolysis in (h),  $V_s$  is the volume of the electrolyte solution in (L), and  $\Delta \text{Conc.}$  is the experimentally measured phenol removed in (mg/L).

From Figures 4 to 6, it is clear that SPC is higher for the cells that use CF as anode and graphite as cathode and lower for the cells that use CF as cathode and graphite as an anode. The highest SPC is achieved after 180 minutes. SPC values of the direct oxidation process were 124.5 and 125.2 (kWh/kg phenol) after 180 minutes when using G-G and G-CF cells (as shown in Figure 4), while SPC reached 1048.4 and 729.2 (kWh/kg phenol) when CF-G and CF-CF electrodes were used. Figure 5 shows that SPC by the indirect electro-oxidation were 76.5, 78.0, 577.8, and 513.5 (kWh/kg phenol) after 180 minutes when using G-G, G-CF, CF-G, and CF-CF. Similarly, Figure 6 shows that SPC after 180 minutes by electro-Fenton oxidation process were 62.5, 57.1, 436.0, and 385.9 (kWh/kg phenol) using the same cells configurations.

The average potential in the direct oxidation process is about 7.5 volt for the arrangement (CF-G) and (CF-CF) and about 7.35 volt for the arrangement (G-G) and (G-CF). So, when the difference in concentrations ( $\Delta \text{Conc.}$ ) are increased for the same current, the SPC decreases. Similarity for the indirect oxidation process, the potential is about 6.9 volt for the arrangement (CF-G) and (CF-CF) and is about 6.5 for the arrangement (G-G) and (G-CF). For the electro-Fenton process, the average potential is about 6.8 volt for the arrangement (CF-G) and (CF-CF) and is about 6.1 arrangement (G-G) and (G-CF).

Table 2 Electro-oxidation rate constants and correlation coefficient of oxidation reaction for different electro-oxidation techniques and various cells configuration

Cells configuration (Anode-Cathode)	Direct		Indirect		Fenton	
	k, min <sup>-1</sup>	R <sup>2</sup>	k, min <sup>-1</sup>	R <sup>2</sup>	k, min <sup>-1</sup>	R <sup>2</sup>
G-G	0.0023	0.9662	0.0038	0.9278	0.0044	0.9286
G-CF	0.0021	0.9638	0.0037	0.9228	0.0054	0.9123
CF-G	0.0004	0.9543	0.0005	0.9093	0.0007	0.9200
CF-CF	0.0003	0.9412	0.0004	0.9040	0.0006	0.9397

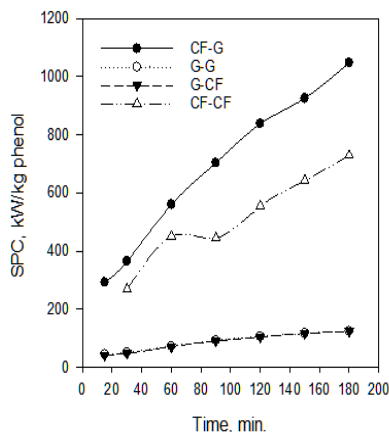


Fig. 4. SPC of phenol removal via direct oxidation process over various cells' arrangements.

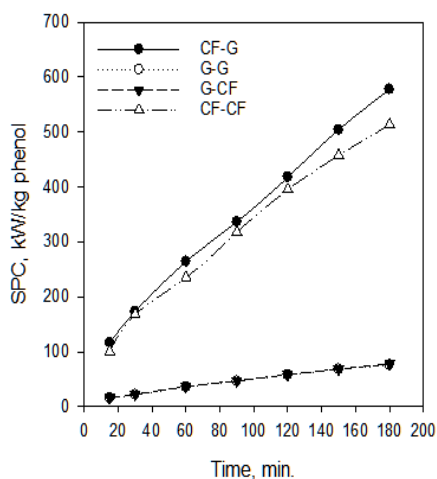


Fig. 5. SPC of phenol removal via indirect oxidation process over various cells' arrangements.

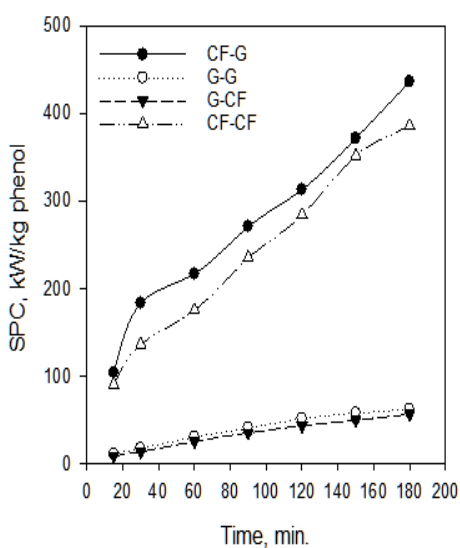


Fig. 6. SPC of phenol removal electro-Fenton oxidation process over various cells configurations.

## Conclusions

Electro-oxidation processes were used to remove phenolic compounds in simulated wastewater with a concentration of 150 ppm by the direct, indirect, and electro-Fenton oxidation processes. The oxidation process is carried by using two-electrode and four cell arrangements. The two electrodes were carbon fiber and graphite, and the four arrangements were (G-G, G-CF, CF-G, and CF-CF). The results of the recorded removal of phenol after 180 minutes using a current density of 8 mA/cm<sup>2</sup> and at a temperature of 35 °C, showed that the performance of the indirect electro-oxidation process is greater than that of the direct electro-oxidation process and that the electro-Fenton oxidation process is the best. The highest phenol removal achieved is about 55.59% by electro-Fenton oxidation technique in the cell arrangement (G-CF). Also, the Fenton process maintained was faster than the other electro-oxidation processes because it has the highest reaction rate constant. Direct oxidation was the slowest process between them because it has the lowest reaction rate constant. The highest performance of the electro-Fenton oxidation process is due to the easy formation of •OH, which is the strongest oxidizer. In contrast to the formation of the chlorine, which contributes to the indirect oxidation process, while the low performance of the direct oxidation process is due to the slow oxidation reaction on the surface of the anode. In the electro-Fenton oxidation process, CF plays an essential role as a cathode because of its high surface area that generates high amounts of hydrogen peroxide that transform to hydroxyl radical in the presence of ferrous ion as a catalyst. Specific power consumption for the electro-Fenton oxidation process is lower than for the indirect oxidation process, while SPC for the direct oxidation process is the highest. Also, that SPC is higher when using CF as an anode and it is equal to 1048.4 and 729.2 (kWh/kg phenol) in the cell arrangements (CF-G) and (CF-CF) in direct oxidation process. While SPC is about 577.8, and 513.5 (kWh/kg phenol) in indirect electrochemical oxidation process and about 436.0, and 385.9 (kWh/kg phenol) in electro-Fenton oxidation process for the same cell arrangements.

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