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Investigating the Potential of Electro-coagulation-Flotation (ECF) Process for Pollutants Removal from Olive Oil Mill Wastewater

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ABSTRACT

Olive oil mill wastewater (OOMW) is a liquid odorous that contains turbidity and organic materials and emulsion oils. More than 30 phenolic compounds were detected in OOMW. The toxicity of OOMW due to phenol compounds is 1000 times more than toxicity of municipal wastewater. Thus, the discharge of wastewater without purification can created problems for the environment. Different methods for OOMW treatment were tested, but none of them is economical and hasn't ideal efficiency. Electrocoagulation–Flotation (ECF) using sacrificial anodes extensively has been used to remove different compounds from water and wastewater. This research is performed on olive oil mill natural wastewater in the laboratorial scale bath electrochemical reactor with 1750 ml useful volume. ECF reactor was consists of an uncovered Plexiglas chamber with pairs of metal sheets called electrodes, that are arranged in pairs of two-anodes and cathodes, the cathode is titanium and anode is commercial iron which they were arranged with a same distance alternately, and they were connected to the direct current power source by parallel monopolar mode. Experiment was performed with 39.06, 78.125 and 117.187 A/m2 current density; in the natural wastewater pH (pH=5.2), neutral pH (pH=7) and alkaline pH (pH=10). The maximum efficiency for all pollutants was obtained at maximum applied current density in the natural pH of wastewater (pH = 5.2). In this manner, by using 117.187 A/m^2 current density in the natural pH of the wastewater after 60 minutes, the efficiency for turbidity, chemical oxygen demand (COD) and phenolic compounds were 99.89%, 96.14%, 89.97% respectively. According to the gained findings, Efficiency treatment electro-coagulation-flotation process for olive oil mill wastewater in natural pH (pH = 5.2), promises the ability this process for olive oil mill wastewater treatment that its pH between 4.5 to 5.5 normally.

KEYWORDS: Olive Oil; Wastewater treatment; COD; Phenol; Electro-coagulation-Flotation.

INTRODUCTION

Industrial wastewaters are the most important contaminant for environment and among them food industries have great share. Olive oil mill wastewater is one of the industrial wastewaters with high volume and for each ton olive fruit about 0.5 to 0.8 m^3 wastewater is produced [1]. Olive oil mill wastewater (OOMW) is a liquid with very unpleasant odor, and very high organic materials and turbidity, and some emulsified oils [2]. Discarding these wastewaters without treatment can cause many problems in the environment [3]. More than 30 phenolic compounds with different density were detected in OOMW and their density was varied about 0.5 – 25 g/l [4]. It was estimated that toxicity of OOMW due to phenol compounds is 1000 times more than toxicity of municipal wastewater [5]. Therefore, uncontrolled discharge of OOMW in the oil producer's countries is a main problem on soil and water environments. Different methods for OOMW treatment were tested such as physical treatment (dilute, sedimentation, centrifuge and filtration), chemical treatment, biological and thermal treatment, but none of them is economical and hasn't ideal efficiency [6 - 9]. Biological methods due to phenolic compounds inhibitor's effect of OOMW on microorganisms are not usable [10].

In the recent decades, electrochemical technique in the developed counties successfully has helped to environment [11, 12]. Electrocoagulation–Flotation (ECF) by sacrificed anode extensively has been used to delete suspension particles, organic compounds, color, metal ions and different compounds from water and wastewater [13-16]. The main mechanism in this process may act as progressive reactions by dynamical processes and may change according to contaminant type and environmental and functional parameters [17, 18]. Many reactions that will occurs in ECF process include forming coagulant ions from anode electrodes, water electrolysis to hydroxyl ion and hydrogen gas by cathode electrodes, floc formation due to reactions coagulant ions and hydroxyl ions an finally flotation pollutants and flocs on the surface solution. Advantages of this process consist of (1) not need to elaborate equipment; (2) easy operation; (3) prevent uses of chemicals; (4) production flocs larger than chemical floc; (5) collected the pollutant on the wastewater by gas bubbles produced during process and finally; (6) production clear and colorless effluent.

The goals of this study are evaluation of ECF process efficiency for pretreatment olive oil mill wastewater, especially, evaluation COD and phenolic compounds removal from olive oil mill wastewater and determination manner of effective factors in efficiency of the process such as pH and current density.

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2. MATERIALS AND METHODS

This research is performed on olive oil mill natural wastewater in the laboratorial scale. According to figure 1 electrochemical reactor was an uncovered Plexiglas chamber with 4 electrode by dimensions 8×8×0.2 centimeter (width, length, thickness) and its electrodes include commercial Iron blades as anode and Titanium as Cathode which they were arranged with a same gap alternately (2 centimeters) and they were connected to the direct current power source (DC power – ZHAOXIN 5A-60V) by monopolar mode (Figure 1).

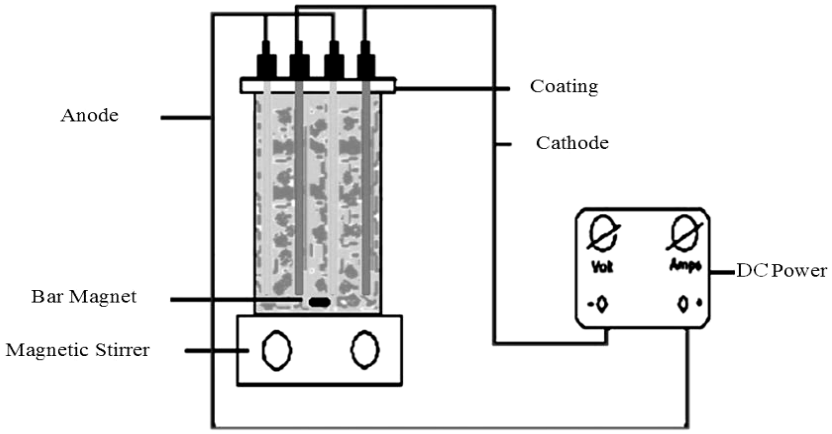

Figure 1. Schematic view of the electrochemical reactor used for ECF process.

In order to create mixing inside of the reactor during the process, a magnetic rod was used, and then the chamber is located on the magnetic mixer with determined round. In each stage of experiment, 1750 ml of wastewater was entered to reactor and by connecting direct current power and creating determined current density (39.06, 78.125 and 117.187 A/m^2); electro-coagulation–flotation (ECF) process in the acidic pH (natural wastewater pH, pH=5.2), neutral pH (pH=7) and alkaline pH (pH=10) was performed. After passage of the determined time (5, 10, 20, 30, 40, 50 and 60 minutes), in each stage a sampling from inside of the reactor was done. Then chemical oxygen demand (COD) and phenol were measured after passing samples through watt man filter size 42. The primary wastewater in each stage was measured by pH- meter WTW inoLab 720. Turbidity of the samples after 30 minutes sedimentation was measured by HACH 2100 N turbidity- meter. Phenol was measured with colorimetric method and 4 - Amino anti-Perrin method by UV- VIS HACH DR 5000 spectrophotometer. In order to measure COD of samples, COD-close reflux method was used. It should be mentioned that all of the contaminants analysis were performed according to the standard methods book [19].

3. RESULTS AND DISCUSSION

3.1 Characteristics of Olive Oil Mill Wastewater

Contaminants of olive oil raw wastewater were measured according to standard methods for water and wastewater tests. Findings are presented in table 1. Figure 2 shows comparison between measured parameters in olive oil mill wastewater and strong municipal wastewater.

Table 1. The basic characteristics of olive oil mill wastewater analyzes

Parameters	Results
pH	5.2
Turbidity$_{(NTU)}$	21400
Total Dissolved Solids (TDS) $_{(mg/L)}$	24900
Chemical Oxygen Demand (COD) $_{(mg/L)}$	36240
Biochemical Oxygen Demand (BOD) $_{(mg/L)}$	11600
Total phenolic compounds$_{(mg/L)}$	299
Chloride $_{(mg/L)}$	1860

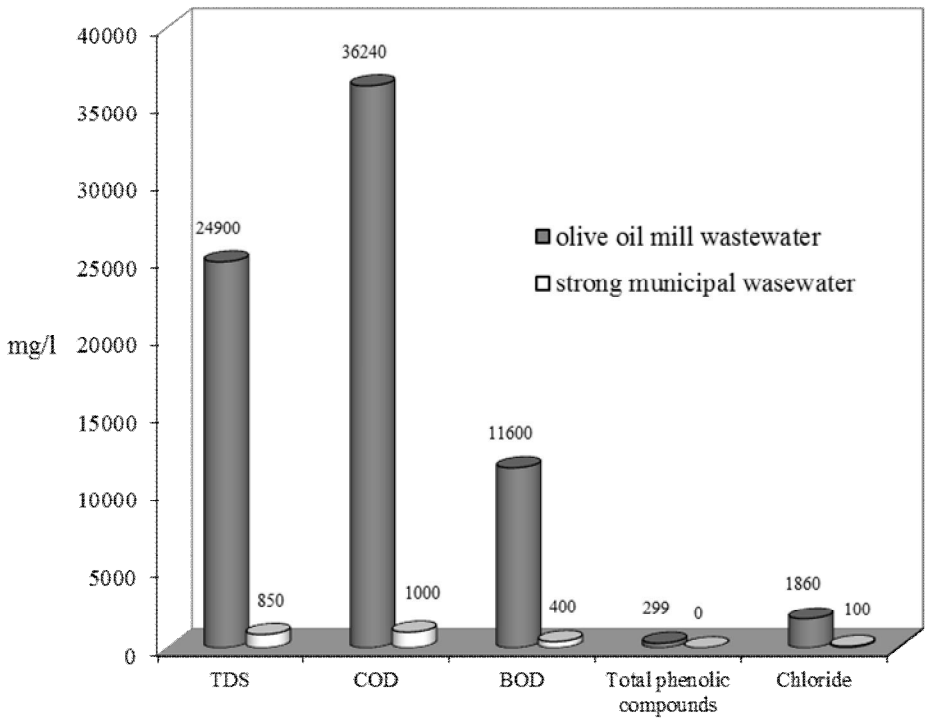

Figure 2. Comparison between measured parameters in OOMW and strong municipal wastewater.

3.2. Influence parameters on the ECF process

3.2.1. pH effect

Studies showed that pH has a key role in efficiency of ECF process. However, in order to study pH effect on ECF process, studies were conducted on three different amounts of pH (pH=5.2, 7 and 10) in current densities from 39.06 to 117.187 A/m^2 and reaction time 5 to 60 minutes. According to results of figures 1 to 3, could be concluded that by increasing pH from 5.2 to 10, efficiency of ECF process in olive oil mill wastewater treatment is decreased. As, pollutants removal efficiency with change situation acidic to basic pH, in 30 minutes with current density of 117.187 A/m^2 , was decreased from 99.76% to 99.67% for turbidity, and from 95.5% to 95.2% for COD and from 88.63% to 86.29% for phenolic compounds. Decreased efficiency for turbidity, COD and phenolic compounds removal in the higher pH is explained by high level of negative charge in the alkaline pH due to anions absorbance from liquid phase to suspension phase. In other word, by increasing pH, the reaction between coagulant's hydroxides of ECF process and the present flocs is decreased [24]. In the basic pH range, coagulant's hydroxide ions due to high level of negative charge, form some ions with negative charge. Goorses and Coworkers in 2002 studied the contaminants removal in the different pHs. Findings showed that increased efficiency of the contaminants removal in pH range of 4 to 7 is depended on forming amorphous hydroxides sedimentation and other hydroxide complexes with hydroxide ions and their polymers [25]. They conclusion that optimum pH for contaminants removal by electro-coagulation is 6. Results of chawaleparet and Angoandi's research in 2009 [26] about ECF process role on biodiesel wastewater treatment, and findings of paracornel Et al research in 2010 [27] about using this process in pretreatment of data oil mill wastewater are in accord with our results. Also, our results are in accord with Adhome and Manser's study, which it reported maximum removal efficiency in COD and Phenol in pH of 4 to 6. According to the olive oil mill natural wastewater pH range (generally between 4.5 to 5.5), can be said that ECF process is usable for olive oil mill wastewater treatment directly and without pH adjustment [28].

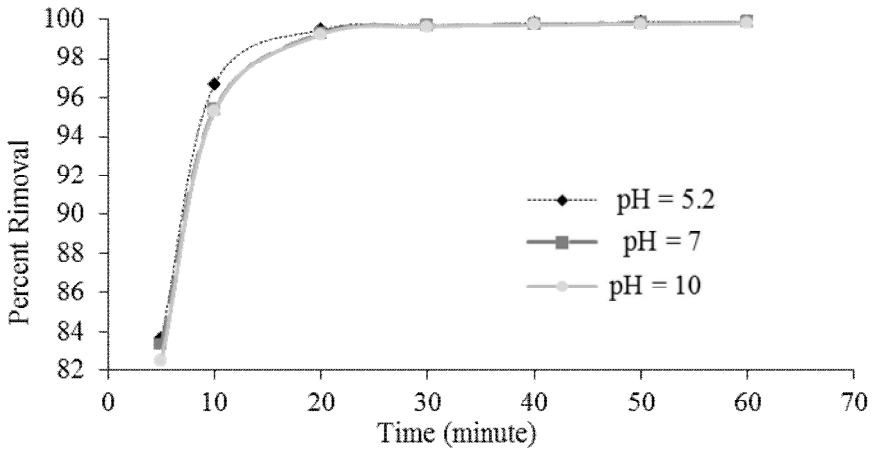

Figure 3. Turbidity removal efficiency of the process in the current density 39.06 A/m^2

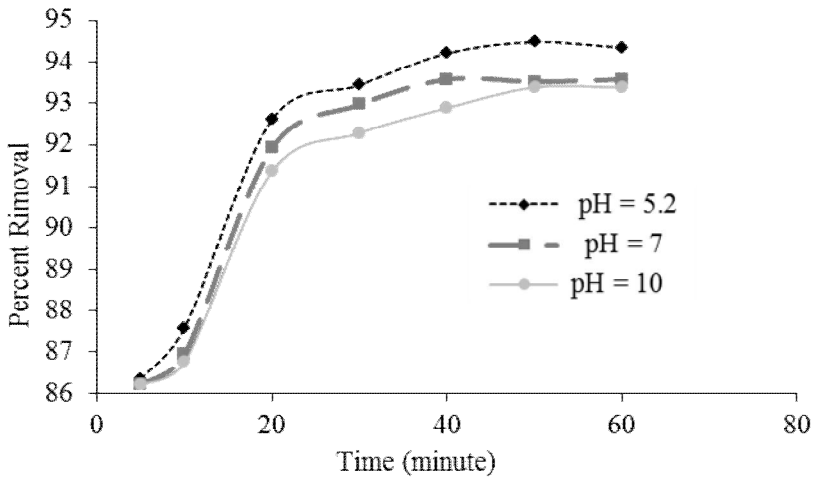

Figure 4. COD removal efficiency of the process in the current density 39.06 A/m^2

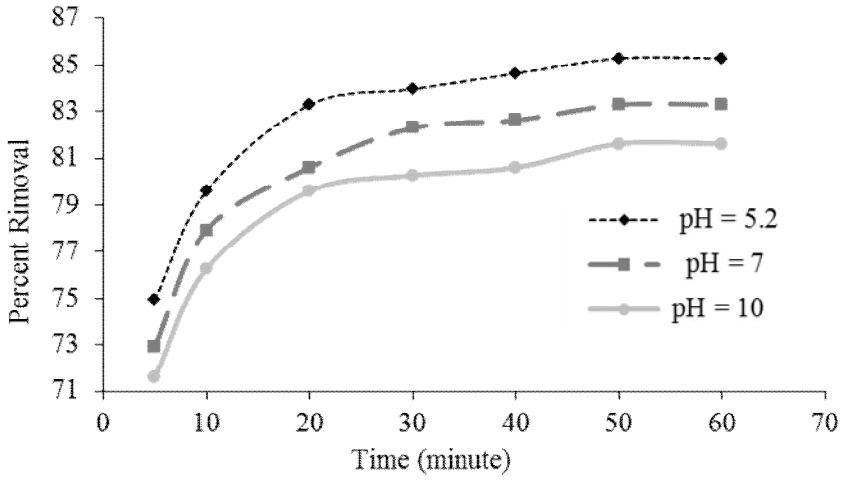

Figure 5. Phenolic compounds removal efficiency of the process in the current density 39.06 A/m^2

3.2.2. Current density effect

Many of the researcher reported that current density can influence on, electro- coagulation and ECF process efficiency directly. In this study, increasing current density from 39.06 to 117.187 A/m^2 in the natural pH of wastewater (pH=5.2) in 60 minutes (reaction time) led to increase in turbidity removal efficiency from 82.48 to 99.89 percent and chemical oxygen demand (COD) from 84.41 to 96.14 percent and phenolic compounds from 73.24 to 89.97 percent. In pH= 7, efficiency of

contaminants removal for turbidity, COD and phenolic compounds were increased from 99.72 to 99.86 percent, 83.77 to 95.86 percent and from 71.57 to 88.96 percent respectively. By increasing current density in PH=10 efficiency of removal process increased from 99.70 to 99.85 percent for turbidity, COD and phenolic compounds respectively. By comparing graphs for a determined countermined contaminant's removal reveals that in the higher current density, the faster is treatment process, and as current density's increased, the necessary time to get the same efficiency is decreased. According to Figure 3, 4 and 5; the maximum efficiency for all of the contaminants' removal in the used current density was gained in the natural pH of wastewater (pH=5.2). Thus, by selecting wastewater's natural pH as the best pH, treatment process in current density's range of 39.06 to 117.187A/m^2 was studied. According to Figure 6, 7 and 8 by using 117.187 A/m^2 current density in the natural pH of the wastewater after 60 minutes, the contaminants removal efficiency for turbidity, COD and phenolic compounds were 99.89%, 96.14% and 89.97% respectively. Also, by increasing potential differences in the electrochemical cell, according to Faraday's law, the amount of the produced coagulant material (used electrode) is increased, and as well the amount of hydroxide flocs is increased and led to more contaminants removal [21, 22].

3.2.3. Process time effect

Reaction time like current density has influence on electrochemical process efficiency. In other word, time and current density have similar effects on ECF efficiency. Efficiency of the contaminant's removal is depended directly on the produced ions concentration of the electrodes. When reaction time increases, ions density is increased too and hydroxide coagulation is formed. However, as graphs 1 to 6 showed, increasing process's time, led to increase in to increase in its efficiency in each amount of current density in the studied pHs. For example, in 117.187 A/ m^2 current density and wastewater natural pH (pH=5.2) changing the reaction time from 5 to 60 min led to increase of efficiency in turbidity removal from 84.36% to 99.89% , COD from 88% to 96.13% and phenolic compounds from 81.61% to 89.97%. By increasing the reaction time from 5 min to 60 min, removal efficiency in pH=7 for turbidity, COD and phenolic compounds are increased from 83.22% to 99.86% , 86.75% to 95.86% and from 79.93% to 88.96% respectively. By increasing the process time in pH=10, the removal efficiency is increased from 82.95% to 99.85%, 86.75% to 98.58% and 73.91% to 87.29% respectively for turbidity, COD and phenolic compounds. In the first 20 minutes of the reaction, especially in the low current density, the amount of Iron cation production in anode is low, so, the amount of produced coagulations and removal efficiency is low too. But by passing time, Iron ions and hydroxyl ions density are increased and the amount of the formed coagulations and removal efficiency of the contaminants are increased too. Also, by increasing electrolysis time along with increased current density, the second reactions may occur such as indirect oxidation of the compounds in presence of cl- ion in the wastewater [23].

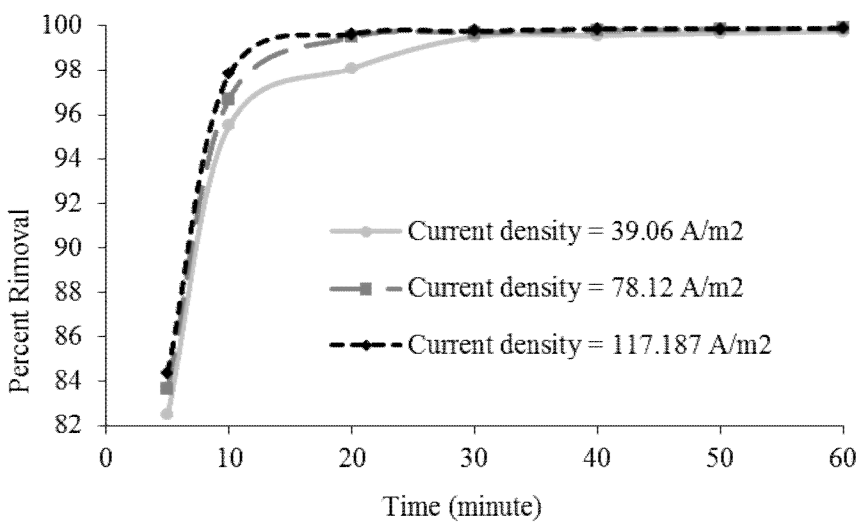

Figure 6. Turbidity removal efficiency of the process in pH 5.2

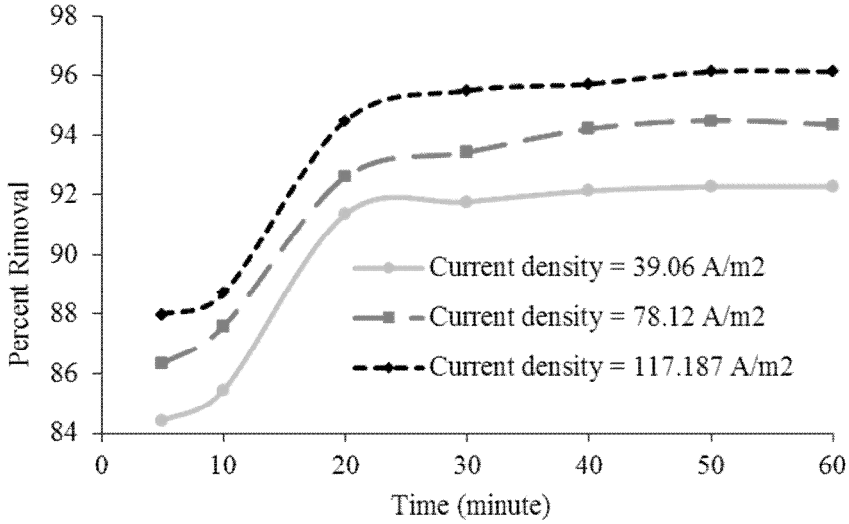

Figure **7**. COD removal efficiency of the process in pH 5.2

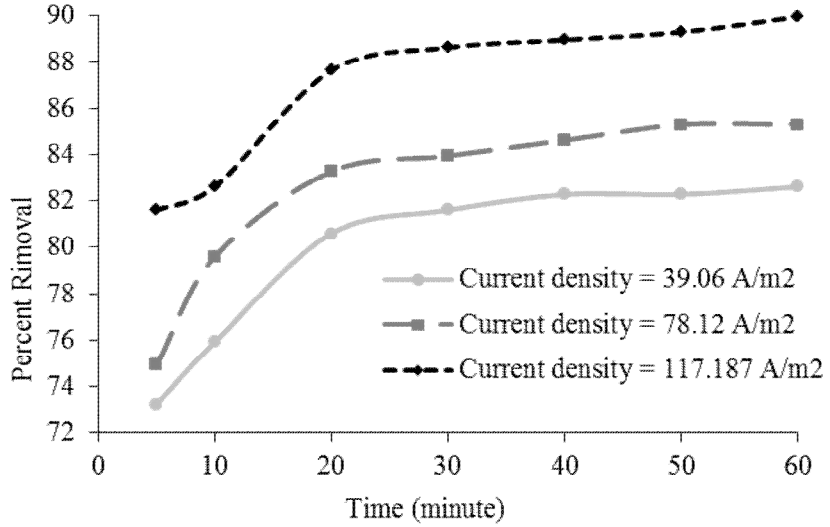

Figure **8**. Phenolic compounds removal efficiency of the process in pH 5.2

4. Conclusion

Research findings showed that load pollution of olive oil mill wastewater with attention to COD is 70 times more than city wastewaters. Also, the amount of phenol in this wastewater is 1000 times more than city wastewaters. In this research, pH effect, current density and process's time were studied for turbidity removal, COD and phenol removal from olive oil mill wastewater by ECF process. Experiment was performed with 39.06, 78.125 and 117.187 A/m2 current density; in the natural wastewater pH (pH=5.2), neutral pH (pH=7) and alkaline pH (pH=10). These experiments showed that the maximum efficiency for all of the contaminants removal by ECF was gained in the natural pH of wastewater (pH=5.2) and highest current density. Decreased efficiency for turbidity, COD and phenolic compounds removal in the higher pH is explained by high level of negative charge in the alkaline pH due to anions absorbance from liquid phase to suspension phase. Efficiency of ECF process in olive oil mill wastewater treatment in the natural pH promising that electro-coagulation–flotation process is usable for olive oil mill wastewater treatment directly and without pH adjustment that its pH range between 4.5 to 5.5.

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