Investigation of sesame processing wastewater treatment with combined electrochemical and membrane processes
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ABSTRACT
This study aims to investigate the treatability of the wastewater generated from the sesame seeds dehulling process by a combination of electrochemical techniques with a membrane filtration system. Chemical oxygen demand (COD) and phenol removal performances were studied for four different cathodes material (iron (Fe), aluminum (Al), platinum (Pt), and boron-doped diamond (BDD)) at different current densities in the electrochemical treatment stage. The maximum removal efficiency was obtained when the BDD electrodes were used. The optimum conditions were 100 A/m² of current density and 120 min of electrolysis period, where 40% and 85% of COD and phenol removals subsequently were achieved. The generated water from the first stage was passed through two different membrane systems. The membrane systems were microfiltration and ultrafiltration. The uptake performance for microfiltration was 22% and 17% for COD and phenol reduction subsequently. The ultrafiltration performed well and has given an additional removal of 27% and 20% of both COD and phenol reduction, respectively. The final results showed the importance of the studied combined systems and the additional value to the final obtained water quality.

Key words | COD, electrochemical process, membrane filtration system, phenol, sesame wastewater

HIGHLIGHTS
- Sesame seeds wastewater treatment has received marginal attention (almost no suggested methods and studies). This study aims to investigate the treatability of the wastewater generated from the sesame seeds dehulling process by combination of electrochemical techniques with membrane filtration system.
- In addition, a study of the effect of cathode type on the removal efficiency of COD and total phenol was conducted.

INTRODUCTION
Sesamum indicum L. or sesame is one of the most common plants, which is cultivated in Asia, Africa, and South America for its seeds (Pham et al. 2010). The plant has different names like Sisim, Till, Ridi, and Benni seeds (Islam et al. 2016). Internationally, it is known as sesame (Deng et al. 2012). According to Bedigian (2010), the genus of the sesame is Sesamum, which contains more than 20 species (Bedigian & Harlan 1986). In general, the plant has a height of 0.5–2 m with a tube shape flower (2–2.5 cm) (Hsu et al. 2011). The fruit (called a seed) has a capsule shape with different colors. Sesame seeds are harvested from the gown plants while the residue parts are thrown, utilized as a soil conditioner, or used as cattle feedstock (Xie et al. 2011). The main chemical seeds compositions are oil with a percentage of 44–57%, protein (18–25%), and 13–14% carbohydrates (Borchani et al. 2010). Sesame seed was the

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first plant that was used as a source of edible oil. Thus, it was mentioned in the literature as the queen of oilseeds (Bedigian & Harlan 1986; Bedigian 2010). Besides that, it has been utilized in domestic, medical, and industrial applications. The harvested seeds are sieved and aspirated to remove any residual dust/stone. Subsequently, dehulling process starts with water injection and sieving. In this process, high amounts of anti-nutritional oxalic acid and fiber are removed from testa (seed coat) (Elleuch et al. 2011).

The dehulling process generates organic waste, inorganic residue, and wastewater (Ngoie et al. 2020). The generated wastewater from the sesame seeds pretreatment process is characterized by high chemical oxygen demand (COD), total solids (TS), oil and greases, and total phenols (Nweke et al. 2014). The discharging of these types into the environment without any treatment causes damages to the aquatic life (Lee et al. 2019), pollutes the ground and surface water (Ma et al. 2015), and odor nuisance (Mar et al. 2016).

The treatment of wastewater by the electrochemical methods (EC) had the researcher’s attention. Electrochemical processes are effective in terms of time, operation, and cost perspectives (Sharma & Simsek 2019). Furthermore, ECs produce low amounts of sludge, require simple devices, and no necessity to use chemical additives (An et al. 2017). The utilizing of membrane systems in wastewater treatment had become favorable. The membrane systems don’t need large areas and can eliminate many kinds of pollutants (Pulido 2016). The main advantages of membrane separation consist of the high efficiency and the continuity of the separation process. However, they do not end in the speed of the process (Seres et al. 2016). The main disadvantages of membranes are fouling aspects, which reduce permeate flux and cause damages to the membrane (Iskandar et al. 2018).

As found in the literature, sesame seeds wastewater treatment has gained marginal attention (almost no suggested methods and studies). This study aims to investigate the treatability of the wastewater generated from the sesame seeds dehulling process by a combination of electrochemical techniques with a membrane filtration system. Also, a study of the effect of cathode type on the removal efficiency of COD and total phenol was conducted.

**MATERIAL AND METHOD**

**Wastewater characterization**

Sesame processing wastewater (SPW) was obtained from a sesame processing factory in Turkey. Wastewater samples were stored at 4 °C until the end of the experiments. For wastewater characterization, COD, total phenol (TP), conductivity, total suspended solids (TSS), total dissolved solids (TDS), and pH were measured according to the American public health association standard methods (Rice et al. 2017). The characteristics of the studied wastewater are shown in Table 1.

**Electrochemical experiments**

EC experiments of sesame processing wastewater were performed in a 300 mL cylindrical borosilicate glass reactor with a working volume of 250 mL. Aluminum, iron, platinum, and boron-doped diamond (BDD) electrodes were used to compare COD and TP removal efficiencies. All electrodes were obtained from Mersin industrial zone, Turkey. The dimensions of used electrodes were 6 cm width × 5 cm height × 1 mm thickness. The immersed active surface area of each electrode was 30 cm². The electrodes were connected to a DC power source (AATech ADC-3303D, maximum output of 30 V). Sesame processing wastewater without pretreatment was added into the reactor, where the reaction under pre-arranged conditions took place. Samples were taken from the reactor at time intervals of 0, 30, 60, 90, and 120 min. The collected samples were analyzed in terms of COD, TP, pH, and conductivity. The removal efficiency of each parameter was calculated using Equation (1).

**Removal Efficiency (%)**

\[
\text{Removal Efficiency} = \frac{(\text{Concentration}_{\text{initial}} - \text{Concentration}_{\text{final}})}{\text{Concentration}_{\text{initial}}} \times 100 \tag{1}
\]

Specific energy consumption (SEC, kWh/kg) is defined in Equation (2). As the electrical energy in kilowatt-hour (kWh), which is required to degrade 1 kg COD equivalent contaminant in the polluted water.

**SEC = \frac{P \times t}{V(COD_i - COD_f)}\tag{2}**

**Table 1 | Raw sesame wastewater characterization**

| Parameter          | Measured value | Unit   |
|--------------------|----------------|--------|
| COD                | 10,010 ± 100   | mg/L   |
| Total phenol       | 248 ± 10       | mg/L   |
| Conductivity       | 188 ± 2        | mS/cm  |
| Total suspended (TSS) | 17,245 ± 525   | mg/L   |
| Total dissolved (TDS) | 10,168 ± 248   | mg/L   |
| pH                 | 6.2 ± 0.02     | –      |
where $P$ is the rated time averaged power (kW), $t$ is the time (h), $V$ is the net treated volume of wastewater (250 mL), COD$_i$ and COD$_f$ are the initial and final COD (kg/L) respectively.

**Membrane experiments**

At the end of the EC experiments, the obtained sesame wastewater was exposed to the filtration experiments. Microfiltration and ultrafiltration experiments were conducted using a dead-end device. The experimental setup consists of a stainless steel cylinder with an effective volume of 300 mL. The inner diameter and the active area were 5.43 cm and 14.6 cm$^2$ correspondingly. The dead-end filtration experiments were carried under a transmembrane pressure (TMP) of 0.1 MPa (1 bar) provided by nitrogen gas. The permeate fluxes were calculated for the microfiltration and ultrafiltration experiments after compaction effects eliminations.

**RESULT AND DISCUSSION**

The electrochemical treatment followed by membranes filtrations were selected as the train for sesame wastewater treatment. For the electrochemical process, four types of electrodes were examined to reach maximum efficiency. Aluminum and iron electrodes were used for the electrocoagulation process realization. Platinum (Pt) and boron-doped diamond (BDD) electrodes were utilized in the electrooxidation process. Both microfiltration and ultrafiltration were studied in the membrane, and the results are illustrated below.

**Fe/Fe electrode pairs**

The electrocoagulation experiment occurred via iron cathodes for 2 h at different current densities (25–150 A/m$^2$) at the original pH. COD, phenol, pH, and conductivity...
changes were tracked over experiment durations. The removals of COD and phenol by the iron (Fe) electrodes have similar behaviour. For COD removal, the increases in the current density increased the removal efficiency from 3.6% at 25 A/m² to 26.7% at 150 A/m² (Figure 1(a)). COD removal increased gradually during the experiment duration and slightly increased by increasing current density. However, phenol removal showed scarcely increase for the 30 min of reaction; after this time, the removal efficiency trend increased rapidly (Figure 1(b)). This effect was also observed during electrochemical treatment of cattle-slaughterhouse wastewater using iron electrodes (Un et al. 2009). In the same manner, the removal efficiency of phenol reached 75.6% at 150 A/m². The supply of current to the electrocoagulation system defines the number of metal ions (Fe²⁺) released from the corresponding electrode. The quantity of resulting coagulant. Therefore, more Fe²⁺ ions get dissolved into the treated wastewater. Current density did not affect pH significantly, as shown in Figure 1(c), where pH remained in the range between 6 and 7 during all the experiment duration. A similar effect was known at conductivity progress, where conductivity did not know many variations. However, it has been known a slight decline with the increase of current density (Figure 1(d)). Similar results were obtained previously (Chandraker et al. 2021). Vasudevan (2014) utilized mild steel as anode and graphite as cathode for phenol removal and the removal efficiency reached 92% at a current density of 0.10 A/dm² and pH of 2.0 (Vasudevan 2014).

**Al/Al electrode pairs**

The electrocoagulation experiments via Al-electrodes were conducted the same as the previous experiment. Sesame wastewater treatment results obtained by Al-cathode are illustrated in Figure 2. Likewise, the ion generation
efficiency increased with the current density increases, which lead to flock production augmentation (Nasrullah et al. 2014). During the first 60 min of the experiment, COD removal efficiency was largely dependent on current density, as demonstrated in Figure 2(a). The maximum removal efficiency was 37%, which is better than the results obtained while used Fe-cathode. Figure 2(b) shows the evolution of phenol removal with time progress and current density. The highest phenol removal performance, which is about 85%, was obtained after 2 h at 150 A/m² of current density. The results also show that the pH range was between 6 and 7 at all the current density over the whole time (Figure 2(c)). Conductivity did not change extensively. However, a slight decline with the increase of current was noticed (Figure 2(d)). The potential differences between the Fe and Al electrodes explain the enhancement in the removal efficiency. The low hydrogen overvoltage of aluminium allowed more pollutants elimination (Benhadji et al. 2011). Ogando et al. (2019) utilized the aluminium electrodes for phenolic, turbidity and colour presents in sugarcane juice and similar results were obtained (Ogando et al. 2019).

**Pt/Pt electrode pairs**

In this section, sesame wastewater was treated using an electrochemical reactor destabilizing pollutant particles present in the wastewater sample using Pt cathode. Figure 3(a) and 3(b) show the relationship between COD and phenol removal efficiencies and electrolysis time applying different current densities. Accordingly, the removal efficiencies increased with the current density and time increases. However, the maximum removal efficiencies for COD and phenol were 25% and 80%, respectively. It can be seen that platinum cathode affected less COD and phenol removal performances compared to an aluminum one. For
pH and conductivity, the obtained results had different trends than other materials (Figure 3(c) and 3(d)). As the time increased, the pH decreased. The main difference while using Pt-cathode is presented in pH variation in function of current density. For 150 A/m², solution pH decreased from +6 to 4 after 2 h of treatment. From Figure 3(d), it is perceived that conductivity is highly influenced and increased by increasing current density opposite what was found in Fe and Al electrodes cases. This can be related to the degradation mechanism which is electrocoagulation in Fe and Al electrodes cases, whereas the main mechanism in Pt/Pt electrodes is electrooxidation (Mebout et al. 1988; Sharma et al. 2019).

BDD/Pt electrode pairs

Figure 4(a) and 4(b) exemplify the evolution of COD and phenol removals, respectively, for the raw sesame wastewater under 120 min of time conditions and different current densities using BDD cathode as experienced with the previously studied cathodes materials. COD removal curves show a sharp tangent and quasi-linear variation in function of time, where up to 40% of COD was reduced, whereas phenol elimination showed similar behavior as much as Al cathode 85%, exceeding results obtained by Fe and Pt cathodes. pH did not change much with the variation of current density as illustrated in Figure 4(c). However, conductivity was affected by current density in particular after 100 min of experiment duration.

Of the four cathodes tested, the efficiency of COD and phenol removal followed the order: BDD, Al, Pt, and then Fe. The efficiency of pollutant removal depended to a certain degree on current density. BDD cathodes are largely used for different wastewater treatments because of their performances (Schmalz et al. 2009; Cruz-González et al. 2010; Frontistis et al. 2011; García-Montoya et al. 2015).
They have always been known for their best performance to remove pollutant which cannot be removed by other treatment processes. For all the studied cathodes material, results of 100 A/m² and 150 A/m² after 120 min of electrolysis were very close and have known relatively the same removal efficiency for both COD and phenol.

Membrane filtration process effect

The effluent of each electrochemical treatment at 100 A/m² of current density was passed through microfiltration (Figure 5(a) and 5(b)) and ultrafiltration systems (Figure 5(c) and 5(d)) at 1 bar of transmembrane pressure. Then, the obtained water was treated and checked for how much membrane filtration enhanced water quality in terms of COD and phenol removal. Both filtration systems have highly improved each of COD and phenol removal of the pretreated sesame wastewater. However, ultrafiltration has known a significant advantage, where it enhanced from 15% to 27% COD elimination and from 12% to 20% phenol removal efficiency. The best train was found with BDD/Pt electrode pretreatment combined with an ultrafiltration system. At the optimum conditions, COD removal was improved by 27% and phenol removal performance increased by 20%. Compared to reported previous studies, ultrafiltration membrane system has always performed better than microfiltration membrane system in terms of COD removal (Cheryan 1998; Muthukumaran & Baskaran 2014; Bouchareb et al. 2020). In this study, an additional pollutant is added to the ultrafiltration membrane removal list, where phenol removal performance was improved for all the treated water samples.

CONCLUSION

This study aims to investigate the treatability of the wastewater generated from the sesame seeds dehulling process.
by a combination of electrochemical techniques with a membrane filtration system. The selected electrochemical process with membrane system is still in its infancy stage, with few research papers published during the last decade. The treatment of sesame industrial wastewater and the removal of emerging contaminants, COD, and phenol have been explored. There are several papers already published about the effective removal of these pollutants by either electrochemical process or membrane filtration process but not the combined process. Moreover, this research was enriched by the study of the cathode material effect on COD and phenol elimination as pretreatment stage. The BOD cathode was proved to be the most efficient for sesame wastewater treatment with the electrochemical process. Furthermore, it was found that ultrafiltration has given better water quality improvement at the same operating conditions compared to microfiltration membrane systems as the second stage of treatment.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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