Turbidity Removal by Electrooxidation from Pistachio Processing Wastewater Using Ti/Pt Anodes

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Summary: Turbidity is the presence of suspended substances in water and is a pollution parameter for water. In this study, the removal of turbidity from pistachio processing wastewater by the electrooxidation process was investigated. Ti/Pt was used as the anode material and stainless steel was used as the cathode material. Stirring speed, supporting electrolyte type, and supporting electrolyte type concentration, initial pH value, and current density were investigated as working parameters. The mixing speed was investigated at 0, 200, 400, and 600 rpm. NaCl, KCl, Na_2SO_4, and $NaNO_3$ were used as supporting electrolyte types. 0, 0.250, 0.500, 0.625, 0.750, and 1.000 M of NaCl concentrations were tested. For pH experiments, 3, 4, 5.3 (natural pH), 7, 9, and 11 values were selected, and studies were carried out at current densities of 5, 7.5, 10, 12.5, and 15 mA/cm². The highest removal rate was found as 66.66% at 400 rpm at mixing speed. The optimum conditions were selected as 0.625 M NaCl electrolyte concentration, 5 mA/cm² current density, wastewater natural pH value (5.3) and turbidity removal was 92.43%, while the energy consumption was 193.5 kWh/m³. As the current density increases, the removal efficiency also increases, but considering the operating cost, this value was chosen as 5 mA/cm². In the results, it was seen that the most effective supporting electrolyte type in the removal of turbidity by electrooxidation was NaCl, the most suitable ambient conditions were the natural pH value of the wastewater, and the electrooxidation was quite efficient.

Keywords: Turbidity Removal**,** Ti/Pt Anode, Electrooxidation, Wastewater Treatment, Energy Consumption.

Introduction

The use and treatment of water, which is an indispensable resource for the continuity of life on Earth, is extremely important. Today, with industrialization and population growth, water consumption is increasing, and the amount of wastewater generated as a result is increasing day by day. Conscious use of water and treatment of used water is the main factor in solving this problem. Physical-chemical processes [1, 2] are effective at removing contaminants. However, because to the substantial amount of chemicals needed for the procedure and the substantial amount of sludge that is produced after treatment, their use is restricted [3]. Once more, biological systems are not always appropriate because of their need for a wide physical space, the sensitivity of particular bacteria to certain chemical compounds, and the lengthy treatment procedures [4]. Researchers have recently demonstrated that electrochemical treatment systems are attractive technologies to minimize or lessen pollution issues as a result of restrictive environmental rules and laws [5, 6]. Electrochemical treatment is one of these processes. One of the first visually striking features of water is turbidity in the water. Turbidity can be considered as a pollution parameter that is undesirable in water and needs to be removed. Thanks to electrochemical technologies, organic and toxic pollutants in wastewater are removed by direct treatment or indirect oxidation with the production of oxidants such as hydroxyl radicals [7]. In direct oxidation, active chlorine species are produced from chloride ions to remove pollutants [8]. Indirect anodic oxidation, pollutants are formed by electron transfer from the anode surface. In direct oxidation is based on the direct oxidation of hydroxyl free radicals and shortens the treatment time [9]. Hydroxyl radicals show significant performance in breaking down organic molecules and microbes [10-12].

Recently industrial developments have resulted in the generation of large amounts of wastewater. Wastewater from these industries contains various heavy metals, turbidity, suspended solids, and sludge volume [13-16]. Turbidity indicates the presence of organic particles such as suspended solids in water and soluble in water. These are factors that can induce microbial resistance as well as having a bad appearance of water [17, 18]. Active chlorine species such as Cl_2, HOCl, and ClO^- (Eqs. 1, 2) can be produced by the direct oxidation of chloride ions on the anode surface or by hydroxyl radicals to yield chlorine (Cl_2). Depending on the pH of the solution, Cl_2 becomes disproportionate in most of the

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solution and forms hypochlorous acid which can give rise to hypochlorite ions [19]. As highlighted by Sirés et al. [20] the use of NaCl as the supporting electrolyte type can form chlorinated oxidation intermediates [21].

The reactions that occur during electro-oxidation are:

Anodic reactions:

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (1)$$

$$6HOCl + 3H_2O \rightarrow 2ClO_3^- + 4Cl^- + 12H^+ + 1.5O_2 + 6e^- \quad (2)$$

$$2H_2O \rightarrow O_2 + 4H^+ + 6e^- \quad (3)$$

Cathodic reactions:

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2 \quad (4)$$

$$OCl^- + H_2O + 2e^- \rightarrow Cl^- + 2OH^- \quad (5)$$

Solution reactions:

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \quad (6)$$

$$HOCl \rightarrow H^+ + OCl^- \quad (7)$$

Common features affecting the electrochemical oxidation process include the types of anode materials and the applied current density [22]. The creation of mediators such reactive oxygen species (•OH), reactive chlorine species (Cl_2 and HClO), and reactive sulfate species (SO_4^-•) can all be affected by the composition of the electrode [23, 24]. In this study, Ti/Pt was used as the active anode.

The aim of this study is to examine the effects of operating conditions such as initial pH, supporting electrolyte, supporting electrolyte concentration, and current density on turbidity removal efficiency. Energy consumption is calculated for all these conditions.

Material and Method

Wastewater Supply and Determination of Characteristics

The wastewater used in the studies was obtained from the output of the pistachio processing plant with an average daily capacity of 24 tons. The wastewater generated in this facility is discharged to the sewer without any physical, biological, or chemical treatment. Afterward, the wastewater was brought to the laboratory in drums, and by using a cooling circulator; the wastewater was kept intact at +4ºC until it was used. In Table-1 the results of the analysis regarding the characterization of this wastewater after it has been left to settle for 6 hours are given.

Table-1: Properties of wastewater.

PARAMETERS	UNIT	VALUES
Conductivity	(µs/cm)	5500
Turbidity	(NTU)	177
pH	-	5.3
COD	(mg/L)	22000

Experimental

A 2000 mL reactor made of fiberglass was used in the electrooxidation experiments. A Chroma brand digitally controlled direct current power supply (62024P–40–120 model 0-40V, 1-120A) was used to provide the required electrical current during the trials. To ensure a good homogenization of the mixing unit, a digital magnetic stirrer (Heidolph MR-3004) was installed in the system with a WTW brand multimeter to be able to adjust the pH, conductivity, and temperature values of the wastewater at the beginning of the reaction and to read these values instantly during the reaction. Plates with dimensions of 70 mm x 100 mm were used. The electrodes have a total immersed area of 2000 cm^2. The distance between the plates was chosen as 5 mm and a total of 10 plates (5 anodes and 5 cathodes) were used. Electricity was supplied to the system using a direct current power supply, and the solution was stirred continuously with the help of a magnetic stirrer. Stainless steel was used as the cathode material and Ti/Pt anodes were used as the anode materials.

Analytical methods

Different parameters have been studied to examine the removal of turbidity from PPIW and how changing conditions in this treatment affect the yield. The turbidity of the water samples was determined using a turbidimeter and expressed as nephelometric turbidity units (NTU) [25]. For analysis, 10 mL of sample was taken from approximately 5 cm below the water surface. Direct electric current passes through both electrodes in the aquatic environment and causes a series of redox reactions in the water and changes in the surface of both electrodes [26]. The turbidity removal efficiency is calculated as follows:

$$T_R = \frac{T_i - T_f}{T_i} \times 100 \quad (8)$$

where, T_i and T_f represent initial and final turbidity (NTU), respectively

Energy consumption (EC):

$$EC = \frac{V \times I \times t}{v} \quad (9)$$

where EC (kW-h/m^3) is energy consumption, V (Volt) is the potential difference, I (ampere) is the current density, t (hour) is the time, and v (m^3) is PPIW the volume.

Results and Discussion

Effects of mixing speed

The effect of mixing speed on turbidity removal was investigated at the natural pH value of wastewater, the current density of 5 mA/cm^2, without using SE. In the results obtained, it was seen that the highest efficiency was at 400 rpm. The turbidity yield was 63.41% without mixing speed, compared to 66.66% at 400 rpm. From the results, it was seen that the mixing speed was not very effective in turbidity removal. Therefore, it was preferred not to use mixing speed to avoid extra energy consumption in studies on other parameters [27]. The results are shown in Fig 2.

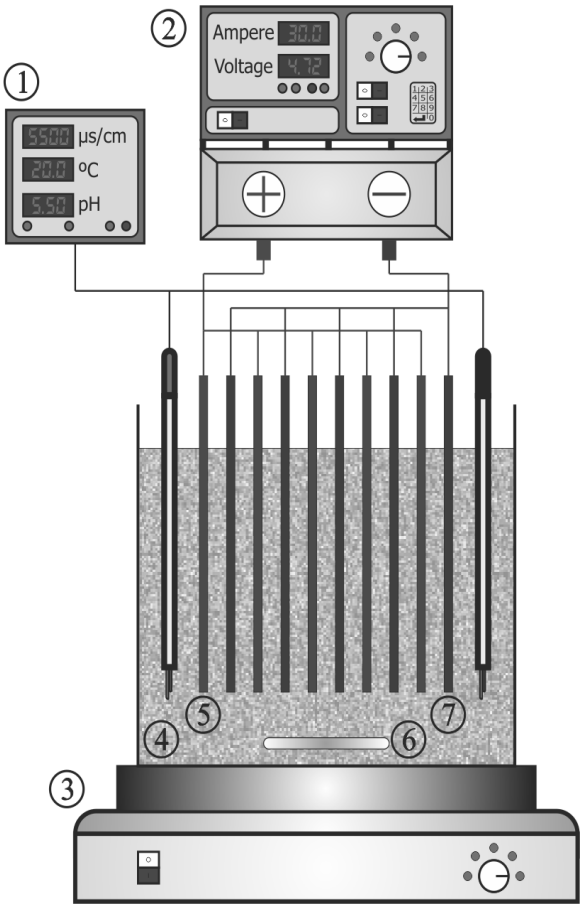

Fig. 1: Electrooxidation reactor setup (1- Multiparameter, 2- DC Power supply, 3- Magnetich stirrer, 4- Wastewater, 5-Ti/Pt anode, 6- Magnet, 7- Steel cathode).

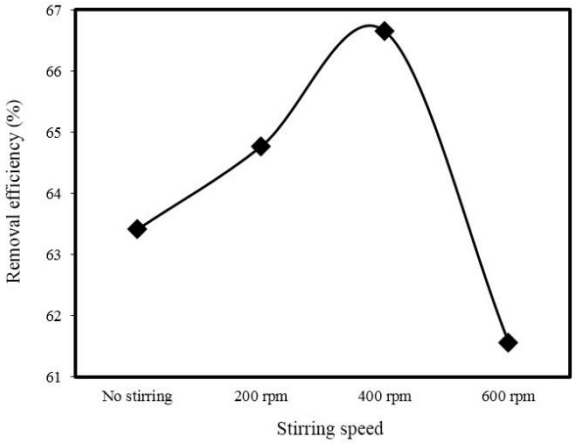

Fig. 2: Effect of mixing speed on turbidity removal turbidity removal (5 mA/cm^2 CD, natural pH, without SE).

Effect of SE type

In electrochemical processes, the type of SE is an important parameter for the removal. Because the added electrolytes affect the water environment and create a suitable environment for oxidation. The working conditions were carried out at a CD of 5 mA/cm^2, at the natural pH value of the wastewater, with a reaction time of 3 hours without mixing speed. In the results obtained, the efficiency was found to be NaCl>KCI>$NaNO_3$>Na_2SO_4>Without SE, respectively. The numerical values of the efficiencies are 83.97%, 78.82%, 71.08%, 68.29%, and 63.41%, respectively. The results are shown in Fig 3. The addition of electrolyte salt in the EO process is highly effective in removing organic material [28]. This gives a higher removal performance to solutions containing NaCl [29]. The reason why the highest efficiency is obtained in NaCl is that it creates various oxidizers, such as hypochlorite and chlorine gas, in the environment due to its high ionization power and chlorine content. The most efficient results in NaCl have been supported by other similar studies [29]. In addition, Chiang et al. [30] among sulfate, nitrate, and chloride, it was reported that the best SE for the electrochemical oxidation of organic pollutants is chloride. Similarly, NaCl was found to be the most effective electrolyte in the study by Govindaraj et al. [31].

Effect of SEC

Studies to examine the effect of SEC were carried out at a current density of 5 mA/cm^2, at natural pH, and at a reaction time of 3 hours without stirring

speed. Studies were carried out at concentrations of 0 M, 0.25 M, 0.5 M, 0.625 M, 0.75 M, and 1 M. In the studies, the optimum SEC was found to be 0.625, and this concentration was studied in the next trials. It was observed that the removal increased as the concentration increased. Because as the added electrolyte increases, the conductivity of the water increases, and it provides higher efficiency in a shorter time. While the efficiency was 63.41% without SE the efficiency increased to 96.0% at 1 M concentration. The reason for this increase is the mediation of active chlorine electrogenerated on the anode surface with the oxidation of chloride given in equations 1.6 and 1.7 [32]. The results are shown in Fig 4.

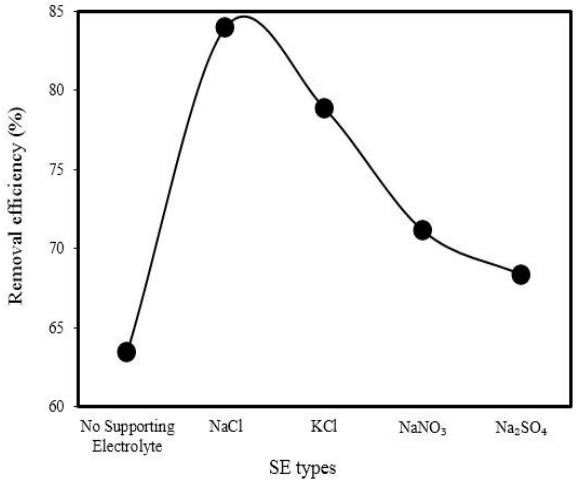

Fig. 3: Effect of SE type on turbidity removal (5 mA/cm^2 CD, 0.5 M, natural pH, without mixing speed).

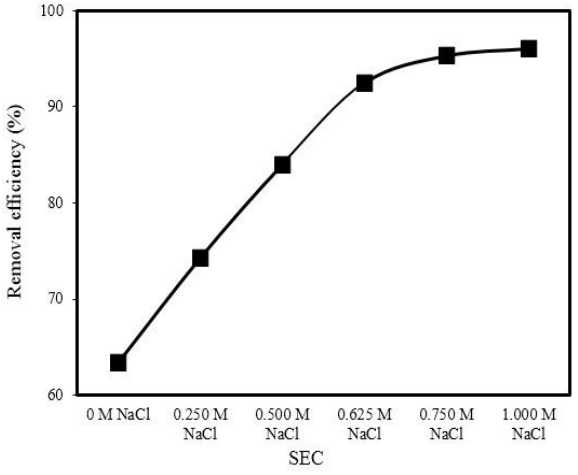

Fig. 4: Effect of SEC on turbidity removal (5 mA/cm^2 CD, natural pH, without mixing speed).

Effect of initial pH

pH is an important operating factor affecting the performance of the EO process [32]. The conditions of the aquatic environment are directly related to process efficiency. In this study, studies were carried out at different pH values such as pH 3.0, 4.0, 5.3 (natural pH), 7.0, 9.0, and 11.0 on turbidity removal the effect of pH value was investigated. The efficiency obtained are as follows for the above mentioned pH values, respectively: 79.60%, 85.93%, 92.43%, 76.59%, 71.64%, and 68.90%. This may be due to reduced chlorine/hypochlorite formation due to chlorate or perchlorate formation at higher pH conditions [32]. The initial pH value often influences indirect oxidation processes and pH controls by the oxidant species produced as mediators in solution (OCl^- at pH> 7, HOCl at pH<5 for NaCl, and $H_2S_2O_8$ in an acidic medium for Na_2SO_4) [33]. As can be seen, the highest turbidity removal efficiency was obtained at the natural pH value of the wastewater. The results are shown in Fig 5.

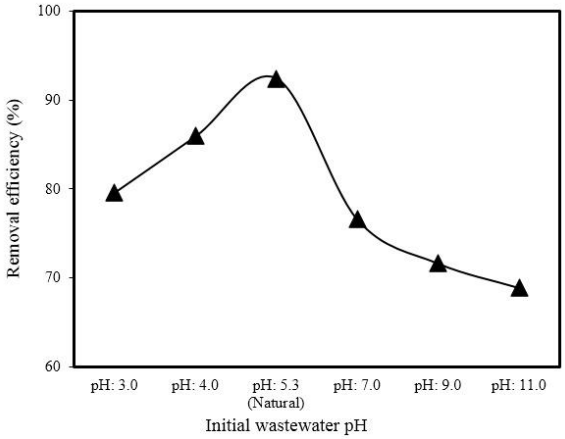

Fig. 5: Initial pH effect on turbidity removal (5 mA/cm^2 CD, 0.625 M NaCl, without mixing speed).

Effect of Current Density

Current density is important in electrochemical processes as it is the driving force in charge migration. It is directly related to the operating cost. In addition, a high current densities will assist in the electrochemical oxidation of organic compounds by promoting the formation of a large OH radical [7]. High current density increases the removal efficiency and shortens the treatment time, as well as increases the operating cost. Therefore, it is important to determine the optimum operating conditions and

reduce the cost. In order to examine the current density effect, studies were carried out at current densities of 5, 7.5, 10, 12.5, and 15 mA/cm². To examine the current density effect, studies were carried out at current densities of 5, 7.5, 10, 12.5, and 15 mA/cm². The obtained turbidity efficiencies were found to be 92.43%, 93.86%, 96.30%, 98.45%, and 99.67% for the specified current densities, respectively. The results are shown in Fig 6. The results showed that the turbidity efficiency increased with increasing current density. As seen in the results, while it was 92.43% at 5 mA/cm^2, it was 99.67% at 15 mA/cm^2 current density. When the current density tripled, there was no significant change in efficiency. Therefore, 5 mA/cm^2 was found to be appropriate for the operating cost.

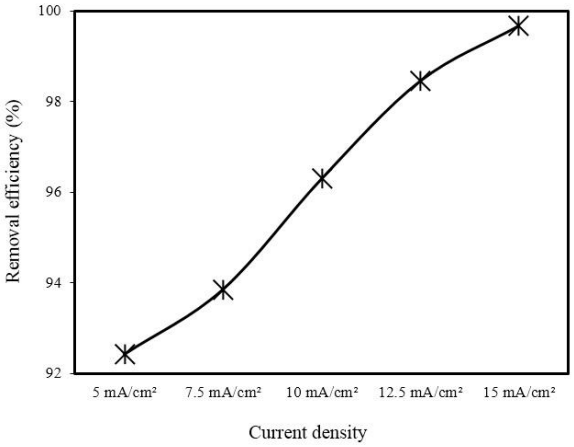

Fig. 6: Effect of CD on turbidity removal (natural pH, 5 mA/cm^2 CD, 0.625 M NaCI, without mixing speed).

Energy Consumptions

Effect of mixing speed on energy consumption

When the effect of mixing speed on energy consumption was examined, it was observed that the energy consumption was the lowest at 400 rpm, where the highest turbidity removal was observed. The energy consumption values obtained at 200, 400, and 600 rpm without mixing speed are as follows, respectively. 289.5 kWh/m^3, 269.5 kWh/m^3, 245.5 kWh/m^3, and 314.5 kWh/m^3. The results are given in Fig. 7.

Effect of SE type on energy consumption

The effect of SE type on energy consumption was investigated for NaCl, KCl, $NaNO_3$, and Na_2SO_4 at the natural pH value of wastewater, current density of 5 mA/cm^2, 0.5 M concentrations for 3 hours reaction time. The results are given in Fig 8. According to these results, it was found 289.5 kWh/m^3 for conditions without SE, 203.5 kWh/m^3 for NaCl, 182.5 kWh/m^3 for KCl, 235.0 kWh/m^3 for $NaNO_3$, and 221.5 kWh/m^3 for Na_2SO_4. The results show that the energy consumption of KCI and NaCl is lower. The reason for this is that these electrolytes contain chloride ions in their structures and therefore they are more active in providing conductivity [34].

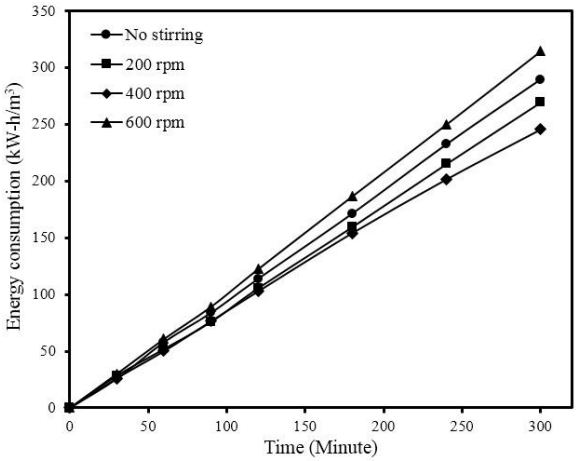

Fig. **7:** Effect of mixing speed on EC (5 mA/cm^2 CD, natural pH, without SE)

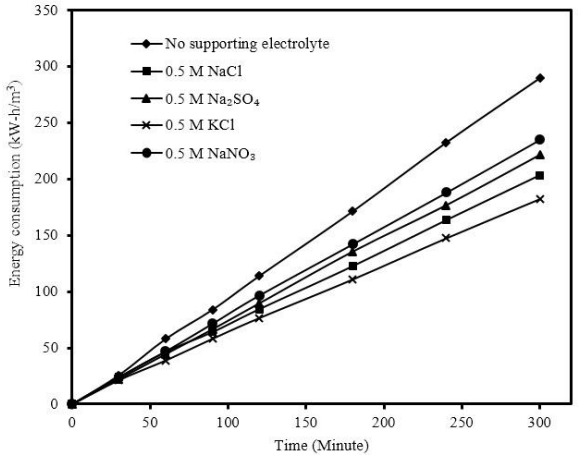

Fig 8**:** Effect of SE type on EC (5 mA/cm^2 CD, 0.5 M, natural pH, without mixing speed).

Effects of SEC on energy consumption

The effect of SEC on EC was investigated using NaCl at concentrations of 0, 0.25, 0.50, 0.625, 0.750, and 1.00 M. The results are respectively: 289.5 kWh/m^3, 227.5 kWh/m^3, 203.5 kWh/m^3, 193.5 kWh/m^3, 184.5 kWh/m^3, and 175.5 kWh/m^3. An increase of 1 M in the concentration of the SE used

resulted in a 114 kWh/m^3 reduction in EC. As can be seen from the results, the increase in the SEC significantly reduced EC. The reason for this is that as the SEC added to the system increases, the conductivity of the wastewater increases and the EC decreases with the decrease of the potential difference in the system [34]. Similar results have been found by other researchers [31]. The results are given in Fig. 9.

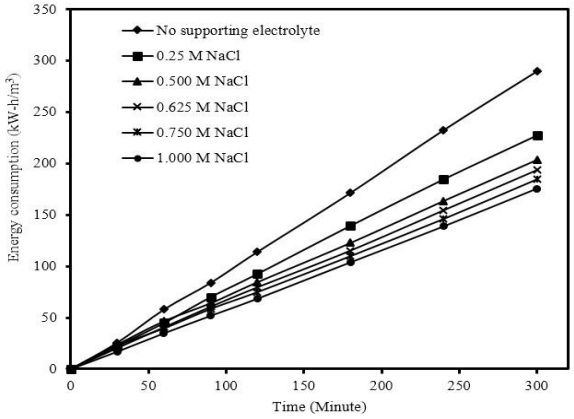

Fig 9**:** Effect of SEC on EC (5 mA/cm^2 CD, natural pH, without mixing speed).

Effect of initial pH value on energy consumption

The effect of the initial pH value of wastewater on energy consumption was investigated at 3.0, 4.0, 5.3 (natural pH), 7.0, 9.0, and 11.0 values, 5 mA/cm^2 current density, and 0.625 M NaCl. In the results obtained, pH: 3.0: 187.0 kWh/m^3, pH: 4.0 188.0 kWh/m^3, pH: 5.3 (natural) 193.5 kWh/m^3, pH: 7.0 178.0 kWh/m^3, pH: 9.0 166.0 kWh/m^3, pH: 11.0 155.0 kWh/m^3 was found. These results are shown in Fig. 10.

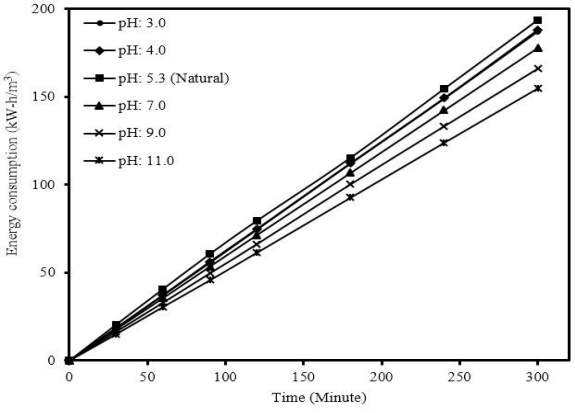

Fig. 10: Effect of initial pH value on EC (5 mA/cm^2 CD, 0.625 M NaCl, without mixing speed).

Effect of current density on energy consumption

The effect of current density on energy consumption, at 0.625 M NaCl concentration, at the natural pH value of wastewater, 5, 7.5, 10, 12.5, and 15 mA/cm^2 current densities were studied. As expected, the energy consumption increased as the current density increased. The energy consumptions for the investigated current densities are as follows: 193.5 kWh/m^3 < 219.0 kWh/m^3 < 247.0 kWh/m^3 < 271.0 kWh/m^3 < 297.5 kWh/m^3. These results are shown in Fig 11.

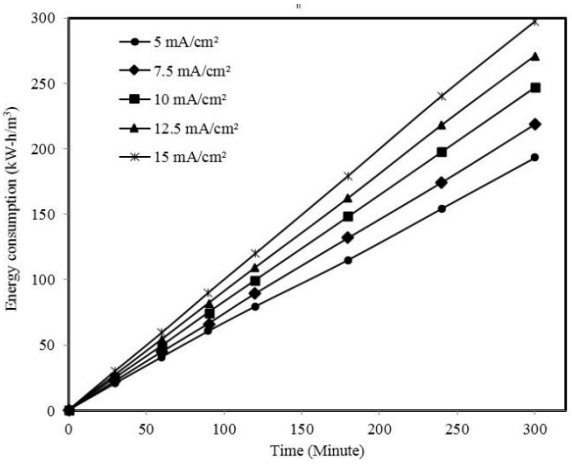

Fig. 11: Effect of CD on EC (natural pH, 5 mA/cm^2 CD, 0.625 M NaCI, without mixing speed).

Conclusions

This present study has shown that the electrooxidation process is quite successful in removing organic matter and turbidity from wastewater. In order to examine the factors affecting turbidity removal from PPIW, studies were carried out under different conditions, and optimum conditions were determined for wastewater. It has been observed that the use of Ti/Pt anode in the removal of turbidity from pistachio processing wastewater provides high efficiency. Among the SE used, it was observed that NaCl was more effective than KCl, Na_2SO_4, and $NaNO_3$. It was observed that the efficiency increased from 74.29% to 96% by increasing the concentration of SE from 0.25 M to 1.0 M. In the results obtained, it was concluded that the most suitable pH environment was the natural pH of the wastewater, and the most suitable current density was 5 mA/cm^2. The yield obtained for these values was 92.43%. In addition, it was observed that the turbidity yield decreased as the pH was increased after the natural pH value. The energy consumption was found to be 193.5 kWh/m^3

under the conditions determined for optimum conditions. For energy consumption, it was observed that NaCl and KCl consumed less energy than other electrolytes, and, energy consumption decreased in basic conditions. It has been concluded that the electrooxidation process can be used as an alternative for large-scale enterprises. In addition, pretreatment by supporting different treatment processes and then using electrochemical processes can increase system efficiency and reduce costs at the same time. In addition to these, the most efficient anode can be preferred by working with different anode materials and comparing the system efficiency.

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Conflict of Interest

The authors have no conflicts of interest to declare that are relevant to the content of this article.

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