

## INVESTIGATION OF TURBIDITY REMOVAL AND ENERGY CONSUMPTION FROM PISTACHIO PROCESSING INDUSTRY WASTEWATER BY ELECTRO-OXIDATION PROCESS

Baybars Ali Fil\* and Sermin Günaslan

Balıkesir University, Engineering Faculty, Department of Environmental Engineering, Balıkesir University, 10146-Balıkesir, Turkey

(Received December 9, 2021; Revised February 22, 2022; Accepted February 22, 2022)

**ABSTRACT.** In this study, it was aimed to remove turbidity, which is an important parameter of water quality, and to examine energy consumption from pistachio processing industry wastewater (PPIW). As working parameters, mixing speed, supporting electrolyte (SE) type and concentration, initial pH value of wastewater, and current density were examined. Graphite was used as the anode material, and stainless steel was used as the cathode material. The highest removal for the mixing speed parameter was achieved at 400 rpm, the maximum efficiency for the SE type was achieved at NaCl, a concentration of 2 M for the SE concentration, a pH of 3.0 for the wastewater initial pH value, and a current density of 30 mA/cm<sup>2</sup>. However, considering the energy consumption cost in these parameters, the optimal conditions were determined as 1.25 M of SE, pH 3.0, mixing speed of 400 rpm, and current density of 6 mA/cm<sup>2</sup>. In these conditions, the turbidity removal efficiency was found to be 90.61%. Energy consumption for the same conditions is calculated at 180 kWh/m<sup>3</sup>. In the results obtained, it is observed that energy consumption decreases as the concentration of SE increases. NaCl, which is the electrolyte that provides maximum efficiency in removing turbidity, is also the electrolyte that provides minimum energy consumption. In electrochemical systems, the cost is as important as lifting efficiency. Therefore, the choice should be made, considering which of these two parameters will be the priority.

**KEY WORDS:** Electro-oxidation, Turbidity removal, Graphite anode, Wastewater treatment

### INTRODUCTION

The world's population, which has continued to grow since the existence of humankind, has caused a variety of environmental problems through industrialization and the increase in consumption that it brings. One of them is water pollution. Water is an essential ingredient that is indispensable for all life, for this reason, the conscious use of water and the treatment and recovery of used water is very important. In particular, industrial waters can cause undesirable problems in the aquatic environment by polluting these limited water resources. Industrialization, economic developments and population growth also increase the volume of wastewater generated worldwide. Moreover, it is stated in the United Nations World Water Development Report that more than 80% of the world's wastewater is discharged to the natural environment without treatment [1]. Turbidity creates a major water treatment problem and is an important indicator of water quality [2].

Turbidity is the presence of various contaminants suspended or dissolved in water. Sand, clay, organic and inorganic substances, various microorganisms cause turbidity in water. Wastewater as a result of production may have various characteristics according to the sectors in which they are produced. The turbidity in wastewater is due to the presence of colloidal substances; and is caused by the adsorption of chemical colloids that will cause the formation of undesirable taste and odor in the water [3, 4]. Therefore, treatment becomes mandatory to be ready to meet global environmental regulation standards before being discharged [5]. There are various techniques used in removing turbidity, suspended solids, dissolved solids and color from water and wastewaters, such as chemical precipitation [6], photocatalysis [6], biological treatment [7],

\*Corresponding author. E-mail: baybarsalifil2@gmail.com

This work is licensed under the Creative Commons Attribution 4.0 International License

membrane filtration [8], ion exchange resin [9], coagulation/flocculation [10], and adsorption [11]. Electrochemical treatment methods are one of the most efficient methods that can be used to remove turbidity from water.

Electro-oxidation, which is one of the electrochemical treatment methods, is based on the removal of organic substances in water with the help of an insoluble anode. It is preferred because it is easy to use and operate, as well as obtaining high efficiency for turbidity. The electro-oxidation process can occur directly or indirectly. In the direct oxidation process, organic substances adhere to the anode surface and decomposition occurs with anode reactions. Indirect electrochemical oxidation is known as an environmentally friendly technology that can remove stubborn organic matter and eliminate nitrogen species. In indirect electrochemical oxidation, the degradation of pollutants takes place on the anodic surface by a strong oxidizer. Chlorine is usually used as a strong oxidizer resulting from the anodic oxidation of chloride [12].

In this study, turbidity removal and energy consumption from PPIW were investigated using graphite anode material. Different parameters such as mixing speed, support electrolyte type and concentration, wastewater initial pH value, and current density, were studied.

## EXPERIMENTAL

### *Electrochemical reactor set-up*

A 2000 mL reactor made of fiberglass was used for electro-oxidation studies. A Chroma brand digitally controlled direct current power supply (62024P-40-120 model 0-40 V, 1-120 A) was used to supply the necessary electrical current to the system. In order to ensure a good homogenization of the mixing unit, a Heidolph MR-3004 brand digital magnetic stirrer was installed in the system in order to adjust the pH, conductivity, and temperature values of the wastewater at the beginning of the reaction. The approximate surface area of all plate sizes is calculated as 1000 cm<sup>2</sup>. 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 it stirred continuously the solution with the help of the magnetic stirrer. Stainless steel was used as the cathode material, and in the studies, wastewater samples in 1000 mL volumes were used. The experimental setup is shown in Figure 1.

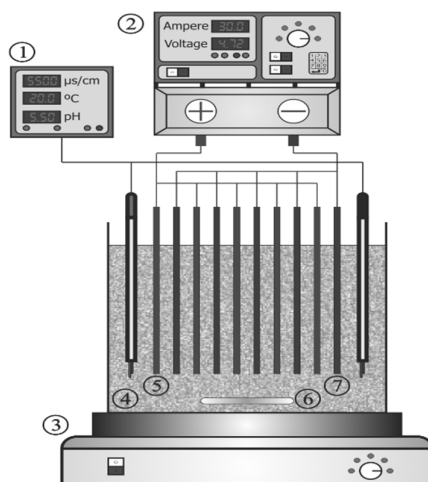


Figure 1. Electro-oxidation reactor set-up.

*Analytical methods*

Different parameters affecting turbidity were investigated in the treatment of PPIW by electro-oxidation method. Turbidity measurements were investigated separately for each experiment. For analysis, 10 mL of sample was taken from approximately 5 cm below the water surface. The turbidity of the water samples was determined using a turbidimeter. Properties of PPIW are shown in the Table 1 [13].

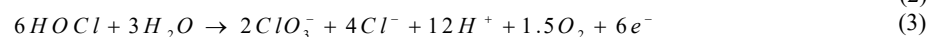
The turbidity removal efficiency is calculated as follows:

$$T_R (\%) = \frac{T_i - T_f}{T_i} \times 100 \quad (1)$$

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

The reactions occurring at the anode and cathode during the process are expressed by the following equations [14].

Anodic reactions:



Cathodic reactions:



Solution reactions:



Table 1. Properties of pistachio processing wastewater.

Parameters	Unit	Values
Conductivity	( $\mu$ s/cm)	5500
Turbidity	(NTU)	177
pH	-	5.3
COD	(mg/L)	22000
TOC (Total Organic)	(mg/L)	5300
TF (Total Phenols)	(mg/L)	4000
Oil-Grease	(mg/L)	55
Antimony (Sb)	( $\mu$ g/L)	<0.046
Arsenic (As)	( $\mu$ g/L)	1
Copper (Cu <sup>+2</sup> )	( $\mu$ g/L)	45
Boron (B)	( $\mu$ g/L)	<0.05
Mercury (Hg)	( $\mu$ g/L)	<0.02
Cadmium (Cd <sup>+2</sup> )	( $\mu$ g/L)	<0.05
Chromium (Cr)	( $\mu$ g/L)	91
Lead (Pb <sup>+2</sup> )	( $\mu$ g/L)	<0.099

## RESULTS AND DISCUSSION

### *Effect of mixing speed on removal efficiency*

In the experiments where graphite anode was used, the effect of mixing speed on turbidity removal efficiency was investigated in 300, 400, and 500 rpm mixing speeds, keeping the pH at 3.0, 6 mA/cm<sup>2</sup> current density, and 1000 mL wastewater sample in the absence of SE. The efficiencies obtained in the experiments were 34.87%, 41.84%, and 39.29%, respectively. It was observed that the highest removal efficiency was obtained at 400 rpm, as seen in Figure 2.

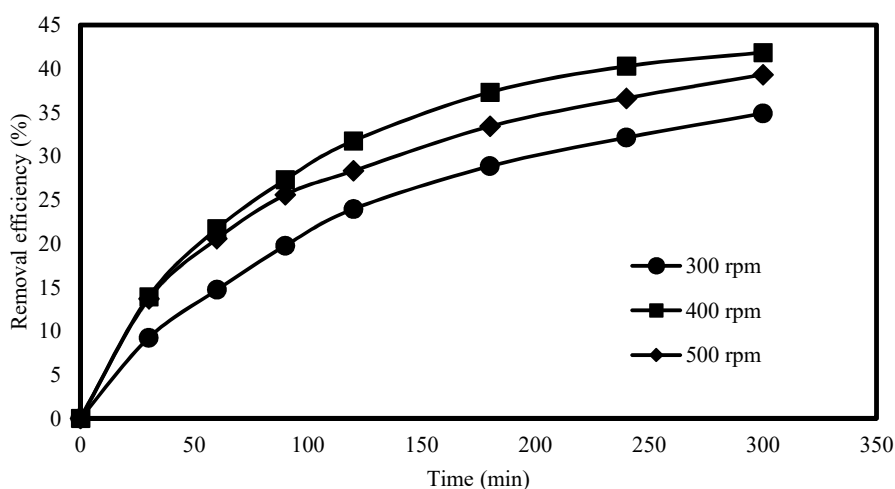


Figure 2. Effect of mixing speed on turbidity removal efficiency.

### *The effect of SE type on turbidity removal efficiency*

While the effect of SE type on turbidity was investigated, the effect of salt types such as NaCl, KCl, Na<sub>2</sub>SO<sub>4</sub> and NaNO<sub>3</sub> were investigated at 1 M concentration, keeping the pH at 3.0, 400 rpm mixing speed and 6 mA/cm<sup>2</sup> current intensity. In the results obtained, it was observed that the highest turbidity removal was the experiment using NaCl electrolyte. The efficiency obtained is arranged in the following decreasing order: NaCl > KCl > Na<sub>2</sub>SO<sub>4</sub> > NaNO<sub>3</sub> > Without SE, respectively. The key factor for the electro-oxidation process is the SE. Because by increasing conductivity, they reduce the energy consumption of the cell, as seen in Figure 3 [15].

### *Effect of SE concentration on turbidity removal efficiency*

In the experiments carried out to investigate the effect of salt concentration on the removal of turbidity; SE concentrations of 0, 0.75, 1, 1.25, 1.5, and 2 M, were studied. Results showed that as the electrolyte concentration increased, the yield also increased. The increase in concentration in the use of SE provides a positive effect in purification. Because the presence of salt in the environment, both conductivity and indirect electro-oxidation can be increased, as seen in Figure 4 [16].

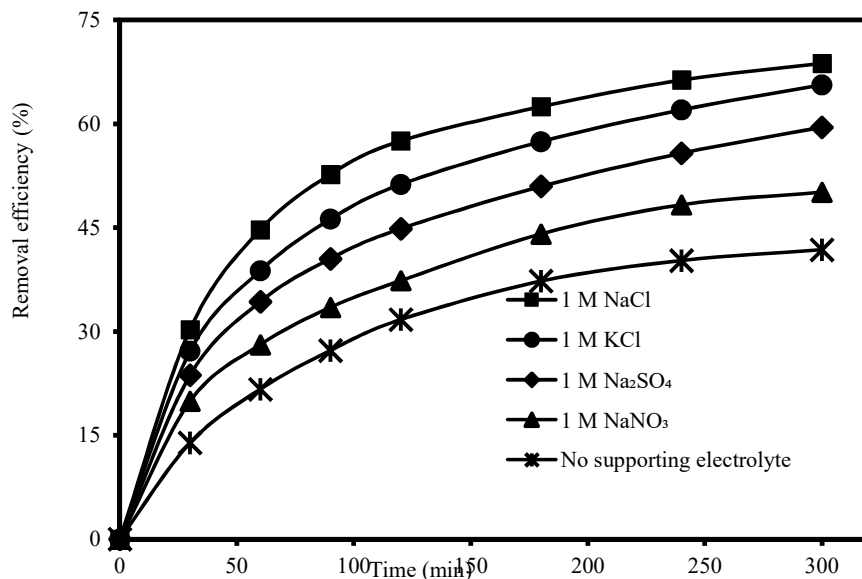


Figure 3. The effect of supporting electrolyte type on turbidity removal efficiency.

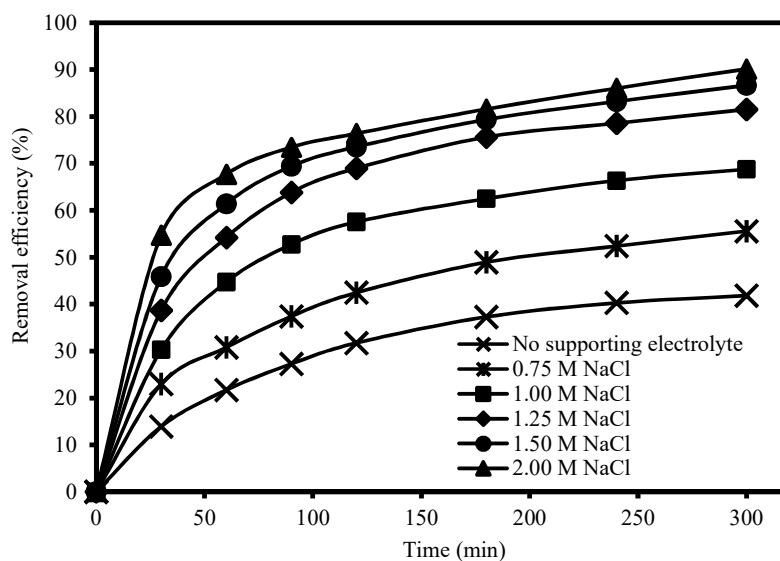


Figure 4. Effect of supporting electrolyte concentration on turbidity removal efficiency.

### *Effect of initial pH value on turbidity removal*

The pH of the water system is always an important parameter which influences the removal of turbidity from the water samples [17]. Wastewater initial pH tests were performed at a SE concentration of 1.25 M NaCl, a current density of 6 mA/cm<sup>2</sup>, and a mixing speed of 400 rpm. Trials were made at different pH values, such as pH 3, 4, 5, 5.3, 6, 7, and 8. The results obtained are 90.61%, 87.20%, 83.60%, 81.53%, 78.11%, 75.74%, and 71.22%, respectively. As can be seen from the results, the highest yield was obtained at pH 3 and the lowest efficiency was obtained at pH 8. From this, it is seen that acidic conditions are more suitable for turbidity removal from peanut processing wastewater, as shown in Figure 5.

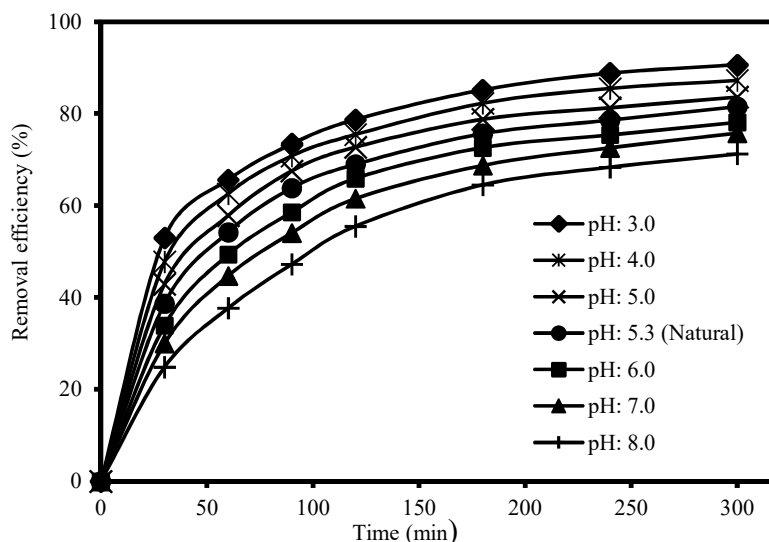


Figure 5. Effect of initial pH value on turbidity removal.

### *The effect of applied current density on turbidity removal*

The current density applied to each unit area of the electrode is preferred as an important parameter in electrochemical processes in terms of controlling the reaction rate [18]. In this part of the study, experiments were carried out at current densities of 6, 10, 14, 18, 22, 26, and 30 mA/cm<sup>2</sup> to see the effect of current density on turbidity removal. As expected, the removal efficiency increased as the current density increased. The main reason here is the higher production of  $\bullet\text{OH}$  and active  $\text{Cl}_2$  species due to the effect of high electron transfer in the medium, which increases the mineralization efficiency of the organic content as seen in Figure 6. [19]. Similar results are available in the literature [20].

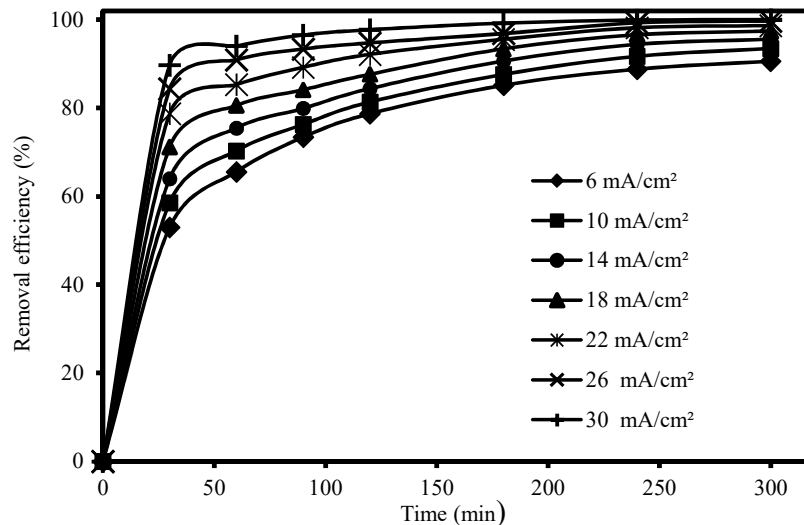


Figure 6. The effect of applied current density on turbidity removal.

#### Energy consumption

Besides the removal of undesirable parameters from wastewater, electricity consumption is also an important factor for these processes because it is directly related to the cost. In all the studies conducted, energy consumption was calculated with the help of the following formula:

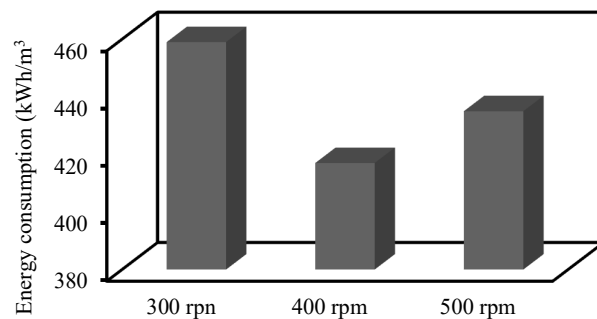
$$\left( \text{kWh} - \text{m}^3 \right) = \frac{V \times I \times t}{v} \quad (9)$$

where, V (volt) is the potential difference, I (ampere) is the current intensity, t (hour) is the time, and v (m<sup>3</sup>) is PPIW the volume.

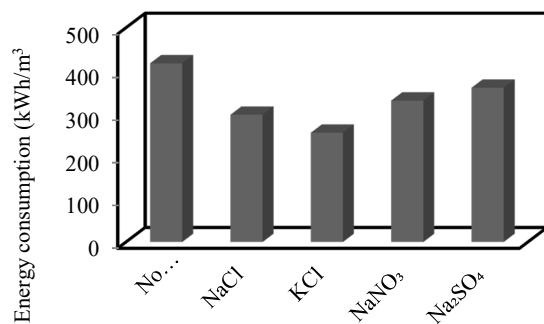
In order to study the effect of mixing speed on energy consumption, studies were carried out on pH 3.0, mixing speed 400 rpm, and current density 6 mA/cm<sup>2</sup>. In experiments examining the effect of mixing speed on energy consumption, 459, 417, and 435 kWh/m<sup>3</sup> were obtained for 300, 400 and 500 rpm, respectively. As can be seen from the results, the lowest energy consumption value was calculated as 417 kWh/m<sup>3</sup> at 400 rpm mixing speed.

When the results for graphite were examined, the lowest energy consumption value was reached at 400 rpm mixing speed. The energy consumption graph is shown in Figure 7a.

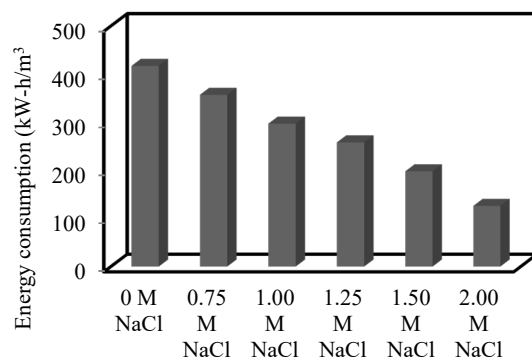
In order to study the effect of different SE on energy consumption, experiments were conducted with NaCl, KCl, NaNO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub> and without SE at the pH 3.0, mixing speed of 400 rpm, current density of 6 mA/cm<sup>2</sup>. When the effect of the SE type on the energy consumption value is examined, it is seen that all trials using the SE consume less energy compared to the ones without SE. The reason for this is that the electrolytes used increase the conductivity of the water and cause a decrease in the potential difference at constant current intensity. Therefore, this means a decrease in electricity consumption as seen in Figure 7b.



(a) stirring speed

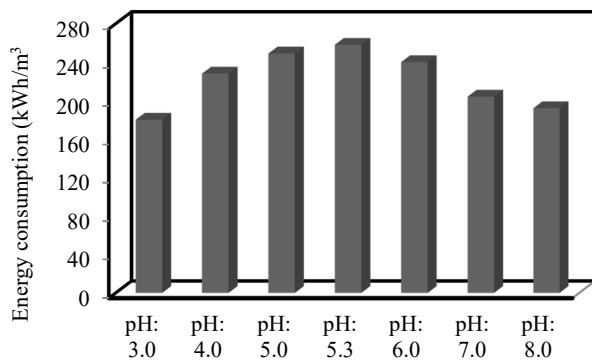


(b) SE types



(c) SE concentration





(d) Initial wastewater pH

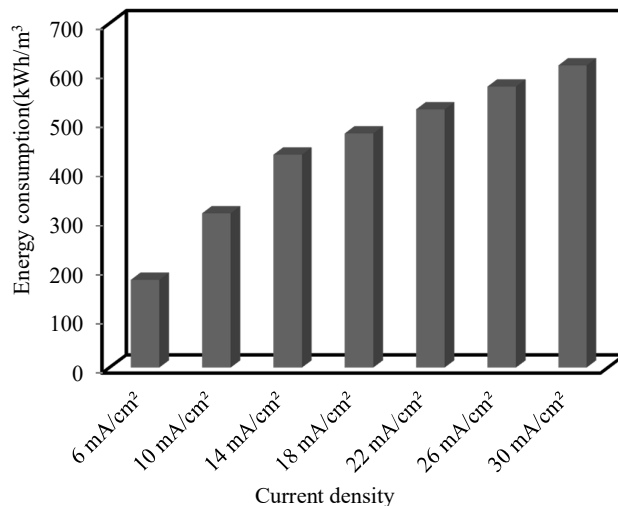


Figure 7. Energy consumption figures.

An increase in the SE concentration means an increase in the electrical conductivity of the solution. When the results are examined, the electricity consumption after 5 hours of operation without SE is 417 kWh/m<sup>3</sup>, while when the SE concentration is 0.75, 1.0, 1.25, 1.5, and 2 M, it is 357, 297, 258, 198, and 126 kWh/m<sup>3</sup>, respectively. In electrolytic cells, solution conductivity affects cell voltage, current efficiency, and electrical energy consumption. Therefore, the electrical conductivity of the solution is an important parameter for saving electricity in an electrochemical process. When the electrical conductivity of the solution is low, it requires more energy to overcome the ohmic resistance between an anode and a cathode, as observed in Figure 7c [21].

The results of the effect of wastewater initial pH on energy consumption are as follows: pH 5.3 > pH 5 > pH 6 > pH 4 > pH 7 > pH 8 > pH 3. Here, it is seen that at the value with the highest removal efficiency (pH 3.0), electricity consumption is also the lowest. In order to examine the effect of pH on electricity consumption, studies were carried out at pH 3.0, 4.0, 5.0, 5.3, 6.0, 7.0,

8.0 and the results are as follows: 180, 228, 249, 258, 240, 204, 192 kWh/m<sup>3</sup>, respectively, as seen in Figure 7d.

The effects of current density on energy consumption were investigated at 6, 10, 14, 18, 22, 26 and 30 mA/cm<sup>2</sup> current densities, keeping the pH at 3.0, 1.25 M SE, and 400 rpm stirring speed in 5-hour trials. In the results obtained, energy consumption was found to be 180, 315, 434, 477, 525.5, 572, and 615 kWh/m<sup>3</sup>, respectively. As can be seen from the results, the lowest energy consumption was achieved at 6 mA/cm<sup>2</sup>; and the highest energy consumption was achieved at 30 mA/cm<sup>2</sup>. Because as the current density increases, the potential difference applied to the system increases, and increasing the potential difference also increases energy consumption [22]. In addition, according to Faraday's Law, increasing the current density also increases the electrode consumption [23]. It is also observed in various studies where high efficiency in turbidity removal has been achieved in similar studies conducted with electrochemical methods as seen in Figure 7e [24-26]. Similarly, it is recommended that very high current densities should not be preferred, since it is seen that energy consumption increases as the current density is increased, and this increases the water temperature [20].

### CONCLUSION

In the present study, turbidity removal and energy consumption from PPIW with the help of graphite anode by electro-oxidation process were investigated. The removal of turbidity in water is aesthetically important. Electrooxidation has also been found to be an effective method for removing water turbidity. It can be preferred due to its ease of use and less sludge formation as a result of the process. From the results obtained, the SE with the highest turbidity efficiency and at the same time the least energy consumption was determined as NaCl. It has been observed that turbidity removal also increases as the SE concentration increases. Increasing the SE concentration increases the conductivity of the medium, allowing the current to flow through the system more easily, and higher efficiency is achieved in a shorter time. The more conductive aquatic environment, the lower the resistance of water pollution to treatment, thus increasing efficiency. Choosing the optimum concentration is important in choosing the electrolyte concentration. The use of high amounts of electrolytes creates both high chemical costs and excessive salinity in the environment. The optimum conditions selected for the operating conditions are determined as 1.25 M NaCl, 6 mA/cm<sup>2</sup> current density, and pH 3.0. In electrochemical processes, cost is as important as efficiency. One of the factors to consider in treatment is cost. It has been observed that turbidity, which is an important indicator of water quality, is effectively removed by electrochemical methods. As expected, the higher the current density, the greater the potential difference in the system, hence the higher the electricity consumption. At this point, efficiency and cost should be calculated and preference should be made according to the priority.

### ACKNOWLEDGEMENTS

The authors are grateful for financial support of Balıkesir University Scientific Research Project Department (Project No: 2019/045).

### REFERENCES

1. Connor, R.; Renata, A.; Ortigara, C.; Koncagül, E.; Uhlenbrook, S.; Lamizana-Diallo, B.M.; Zadeh, S.M.; Qadir, M.; Kjellén, M.; Sjödin, J. The United Nations World Water Development Report. Wastewater: the Untapped Resource. *The United Nations World Water Development Report 2017*.
2. Holmes, T.P. The offsite impact of soil erosion on the water treatment industry. *Land Econ.* **1988**, *64*, 356-366.

3. Okolo, B.; Nnaji, P.; Menkiti, M.; Onukwuli, O. A kinetic investigation of the pulverized okra pod induced coag-flocculation in treatment of paint wastewater. *Am. J. Anal. Chem.* **2015**, *6*, 610-622.
4. Antov, M.G.; Šćiban, M.B.; Prodanović, J.M.; Kukić, D.V.; Vasić, V.M.; Đorđević, T.R.; Milošević, M.M. Common oak (*Quercus robur*) acorn as a source of natural coagulants for water turbidity removal. *Ind. Crops Prod.* **2018**, *117*, 340-346.
5. Okolo, B.I.; Adeyi, O.; Oke, E.O.; Agu, C.M.; Nnaji, P.C.; Akatobi, K.N.; Onukwuli, D.O. Coagulation kinetic study and optimization using response surface methodology for effective removal of turbidity from paint wastewater using natural coagulants. *Sci. Afr.* **2021**, *14*, e00959, 1-25.
6. Huang, M.; Liu, Z.; Li, A.; Yang, H. Dual functionality of a graft starch flocculant: Flocculation and antibacterial performance. *J. Environ. Manage.* **2017**, *196*, 63-71.
7. Yang, G.; Wang, D.; Yang, Q.; Zhao, J.; Liu, Y.; Wang, Q.; Zeng, G.; Li, X.; Li, H. Effect of acetate to glycerol ratio on enhanced biological phosphorus removal. *Chemosphere* **2018**, *196*, 78-86.
8. Furuya, K.; Hafuka, A.; Kuroiwa, M.; Satoh, H.; Watanabe, Y.; Yamamura, H. Development of novel polysulfone membranes with embedded zirconium sulfate-surfactant micelle mesostructure for phosphate recovery from water through membrane filtration. *Water Res.* **2017**, *124*, 521-526.
9. Bui, T.H.; Hong, S.P.; Yoon, J. Development of nanoscale zirconium molybdate embedded anion exchange resin for selective removal of phosphate. *Water Res.* **2018**, *134*, 22-31.
10. Wang, W.; Yue, Q.; Li, R.; Song, W.; Gao, B.; Shen, X. Investigating coagulation behavior of chitosan with different Al species dual-coagulants in dye wastewater treatment. *J. Taiwan Inst. Chem. Eng.* **2017**, *78*, 423-430.
11. Du, Q.; Wang, Y.; Li, A.; Yang, H. Scale-inhibition and flocculation dual-functionality of poly (acrylic acid) grafted starch. *J. Environ. Manage.* **2018**, *210*, 273-279.
12. Nidheesh, P.V.; Kumar, A.; Syam Babu, D.; Scaria, J.; Suresh Kumar, M. Treatment of mixed industrial wastewater by electrocoagulation and indirect electrochemical oxidation. *Chemosphere* **2020**, *251*, 126437.
13. Tırnık, S.; Nuhoğlu, A.; Kul, S. Characterization of pistachio processing industry wastewater and investigation of chemical pretreatment *Environ. Res. Technol.* **2020**, *3*, 209-216.
14. Fil, B.A.; Boncukcuoğlu, R.; Yılmaz, A.E.; Bayar, S. Electro-oxidation of pistachio processing industry wastewater using graphite anode. *Clean* **2014**, *42*, 1232-1238.
15. Abdelhay, A.; Jum'h, I.; Al Bsoul, A.; Arideh, D.; Qatanani, B. Performance of electrochemical oxidation over BDD anode for the treatment of different industrial dye-containing wastewater effluents. *J. Water Reuse Desal.* **2020**, *11*, 110-121.
16. Ghalwa, N.; Abu-Shawish, H.; Tamous, H.; Alharazeen, H. Determination of electrochemical degradation of E102 dye at lead dioxide-doped carbon electrodes using some potentiometric and spectrophotometric methods. *Chem. J.* **2013**, *3*, 1-6.
17. Kumar, P.S.; Vaibhav, K.N.; Rekhi, S.; Thyagarajan, A. Removal of turbidity from washing machine discharge using *Strychnos potatorum* seeds: Parameter optimization and mechanism prediction. *Resour. Efficie. Technol.* **2016**, *2*, S171-S176.
18. Chen, G. Electrochemical technologies in wastewater treatment. *Sep. Purif. Technol.* **2004**, *38*, 11-41.
19. Ken, D.; Sinha, A. Dimensionally stable anode (Ti/RuO<sub>2</sub>) mediated electro-oxidation and multi-response optimization study for remediation of coke-oven wastewater. *J. Environ. Chem. Eng.* **2021**, *9*, 105025.
20. Sanni, I.; Karimi Estahbanati, M.R.; Carabin, A.; Drogui, P. Coupling electrocoagulation with electro-oxidation for COD and phosphorus removal from industrial container wash water. *Sep. Purif. Technol.* **2022**, *282*, 119992.

21. Chou, W.-L.; Wang, C.-T.; Chang, S.-Y. Study of COD and turbidity removal from real oxide-CMP wastewater by iron electrocoagulation and the evaluation of specific energy consumption. *J. Hazard. Mater.* **2009**, 168, 1200-1207.
22. Fil, B.A. *Treatment of Pistachio Processing Wastewater by Electrooxidation Method*. PhD Thesis, Graduate School of Natural and Applied Sciences, Department of Environmental Engineering, Atatürk University, Erzurum, Turkey, **2014**.
23. Kumar, P.; Chaudhari, S.; Khilar, K.; Mahajan, S. Removal of arsenic from water by electrocoagulation. *Chemosphere* **2004**, 55, 1245-1252.
24. Ním, M.; Othman, F.; Sohaili, J.; Fauzia, Z. Removal of COD and turbidity to improve wastewater quality using electrocoagulation technique. *Malaysian J. Anal. Sci.* **2007**, 11, 198-205.
25. Bukhari, A.A. Investigation of the electro-coagulation treatment process for the removal of total suspended solids and turbidity from municipal wastewater. *Bioresour. Technol.* **2008**, 99, 914-921.
26. Ebba, M.; Asaithambi, P.; Alemayehu, E. Investigation on operating parameters and cost using an electrocoagulation process for wastewater treatment. *Appl. Water Sci.* **2021**, 11, 175.