



## Research Article

# Pre-coagulated landfill leachate treatment by synergistic ultraviolet and electro-persulfate process

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## ABSTRACT

In this study, the integration of ultraviolet (UV) and electrochemically activated persulfate (PS) oxidation (UV/Electro-PS) was investigated in pretreated landfill leachate treatment. Optimum process parameters for chemical oxygen demand (COD), UV<sub>254</sub>, and color removal from pre-coagulated landfill leachate were investigated. The process variables and ranges of variables were selected as pH 3-11, applied current 1-5 A, PS dose 1-9 g/L, and reaction time 5-35 min. Optimum process conditions were determined as pH 3, applied current 3 A, PS dose 5 g/L, and reaction time 25 min. By applying optimum conditions, COD, UV<sub>254</sub>, and color removal efficiencies by the UV/Electro-PS process were 55.6%, 68.5%, and 89.9%, respectively. The total energy consumption was calculated as 27.4 kWh/kg COD for the optimum process conditions. UV/Electro-PS process was effective for the COD, UV<sub>254</sub>, and color removal from pre-coagulated landfill leachate.

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## INTRODUCTION

The landfill leachate is formed from the rainwater passing through the waste body in the landfills, the water formed due to the biochemical processes taking place in the landfill storage area, and the water released from the compression of the wastes' liquid content after storage [1]. Leachate can cause major environmental issues by penetrating and combining with soil, lower layers of soil, surface water, and groundwater if not gathered, stored, and treated properly [1]. Since leachate is complex and highly toxic wastewater with high concentrations of organic and

inorganic substances, its treatment is challenging and expensive [2,3]. In the treatment of leachate, more than one process is needed to obtain satisfactory treatment performance. Thus, combined, integrated, or sequential systems give promising results in the efficient treatment of landfill leachate [4,5].

Coagulation can be used as a pretreatment process, especially in sequential systems. The coagulation process is based on the removal of large particles or flocks formed due to the collision of destabilized particles with each other by precipitation, flotation, or rapid filtration mechanisms [6]. Coagulants such as ferric chloride or aluminum

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sulfate are added during traditional chemical coagulation. Furthermore, currently, polymerized metal coagulants like poly aluminum chloride (PAC) have been used as coagulants due to their low cost, reduced sludge buildup, and simple accessibility [7,8].

Advanced oxidation processes (AOPs) are quite successful in removing resistant organic matter in leachate after the organic matter load is reduced by physicochemical methods such as the coagulation process [9–11]. The generation of very reactive free radicals that react with many organic pollutants is the foundation of AOPs. With AOPs, the target pollutant is converted to intermediate and/or final materials that are not as harmful and more effortlessly biodegradable by partial oxidation, or complete mineralization of the pollutant is guaranteed. Hydroxyl radicals and sulfate radicals are involved in advanced oxidation processes [12]. Oxidants widely used in sulfate radical-based AOPs are peroxymonosulfate or peroxydisulfate (PS) [13] and the sulfate radicals can be obtained by activation of these oxidants. Methods such as the temperature increase, electrical current, ultraviolet (UV), the addition of transition metals, and ultrasound can be used singularly or in combination for the activation of oxidants. The hydroxyl radicals, which have a larger redox potential and less specificity, are subsequently formed by the ensuing sulfate radicals [14–16].

There is diverse research on combined coagulation and advanced oxidation process applications. Amor et al. [17] assessed the performance of sequential coagulation/flocculation and Fenton/solar Fenton process for mature leachate treatment, and 75% dissolved organic carbon (DOC) removal was reported with the coagulation/flocculation-solar photo Fenton process. GilPavas et al. [18] used a sequential coagulation-electrooxidation process in textile wastewater treatment and obtained 75% total organic carbon (TOC) and 93.5% chemical oxygen demand (COD) elimination. Tripathy and Kumar [19] investigated leachate treatment with sequential coagulation/flocculation-microwave-activated PS processes and obtained COD removal efficiencies of 73% and 62%, respectively by using  $\text{FeCl}_3$  and alum in pretreatment. Can-Güven et al. [20] reported 82.2% COD removal from leachate with subsequent coagulation and persulfate/peroxide binary oxidation. Umar et al. [21] evaluated the efficiency of the pre-coagulation process in the treatment of domestic wastewater RO concentrate with UVC-LED/ $\text{H}_2\text{O}_2$  and stated that the coagulation, in which  $\text{FeCl}_3$  is applied as a coagulant, provides high performance, especially in DOC removal. Photo/electro/persulfate process applications are very limited in the literature. Ahmadi and Ghanbari [22] applied a photo/electro/persulfate process for greywater treatment and 77% COD removal was obtained. Mehralipour and Kermani [23] evaluated the photo/electro/persulfate/nZVI process performance for dichlorophenoxyacetic acid degradation and reached a removal efficiency of approximately 92% under optimized conditions.

Although there are combined coagulation/advanced oxidation applications in leachate treatment in the literature, to the best of our knowledge, a study including coagulation as the pre-treatment, and the UV/electro/persulfate process as the advanced oxidation process does not exist. In addition, the study contributes significantly to the literature in terms of applying two different activation methods for persulfate oxidation following the coagulation/flocculation process. This study is novel concerning the application of the combined coagulation and UV/Electro-PS process in leachate treatment. In the study, the effects of pH, applied current, PS dose, and reaction time on the UV/Electro-PS process which was applied to pre-coagulated landfill leachate, and the performance of the combined process was investigated.

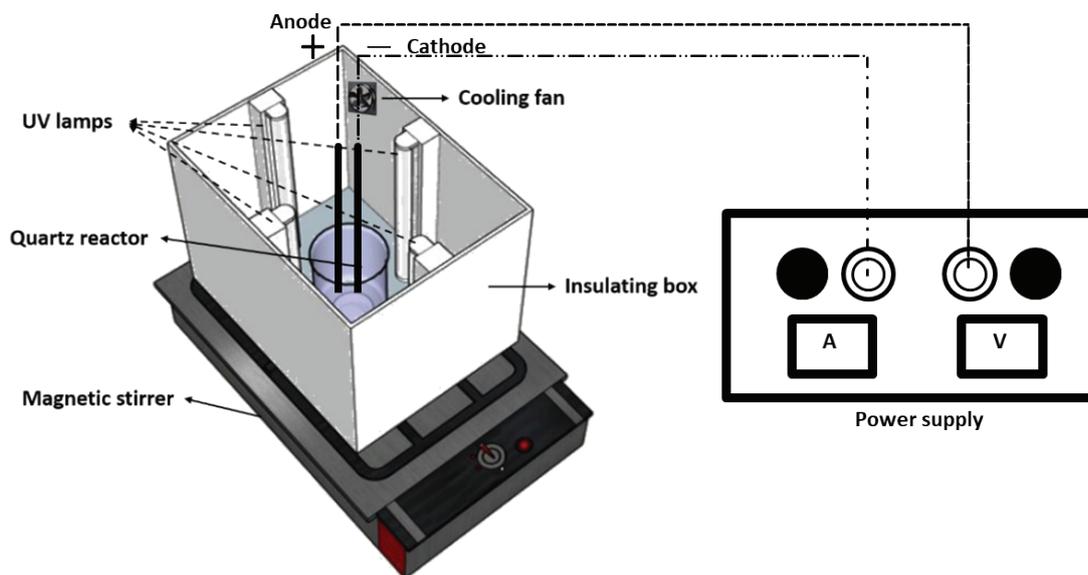
## MATERIALS AND METHODS

### Leachate Characterization and Analytical Methods

The leachate for the experiments was taken from the inlet of the Odayeri Sanitary Landfill Leachate Treatment Plant, Istanbul. Leachate was stored at  $+4^\circ\text{C}$  to preserve the characteristics of the wastewater. The properties of the leachate before and after experimental studies were analyzed by the methods recommended by APHA [24]. WTW Multi 9620 IDS device was utilized for the measurements of the pH and conductivity while WTW-Photolab 6600 UV-Vis spectrophotometer was utilized to determine the color and  $\text{UV}_{254}$  parameters. SM 5220-C and SM 2540-D were used to determine the COD and total suspended solids (TSS) concentrations, respectively. The COD was measured and evaluated to determine the organic matter removal through the treatment processes. High concentrations of humic matter, including humic acid, fulvic acid, and humin exist in leachate and this fraction is resistant to biological treatment. The  $\text{UV}_{254}$  parameter was selected to represent the resistant organic matter fraction. The leachate has a high color content, thus this parameter was chosen in the evaluation of the process performance. The properties of the raw leachate were; pH 8.09, conductivity 37.4 mS/cm, COD 13059 mg/L, TSS 1110 mg/L,  $\text{UV}_{254}$  66.3 1/cm, and color 13150 Pt-Co.

### Coagulation Process

A detailed explanation and discussion of the coagulation were included in our previous study [25]. Briefly, 100 ml of the pH-adjusted leachate was put on the Jar-test apparatus. The specified quantity of PAC was put and mixed at 45 rpm during predetermined reaction times after the rapid agitation at 200 rpm. The impact of PAC dose, coagulation time, and pH on the elimination efficiency of the COD and  $\text{UV}_{254}$  were determined using the Box-Behnken design, and the best operating conditions were determined. In the current study, the effluent of the coagulation of our previous study [25] was used. The properties of the pre-coagulated



**Figure 1.** The schematic display of the UV/Electro-PS process set-up.

leachate used in this study were; pH 9.99, conductivity 37.9 mS/cm, COD 6425 mg/L, TSS 276 mg/L,  $UV_{254}$  25.9 l/cm, and color 2946 Pt-Co.

#### UV/Electro-PS Process

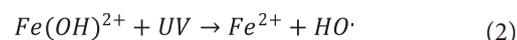
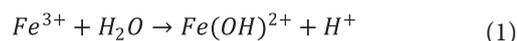
Experimental studies of the UV/Electro-PS process were performed in a 16-cm-high, cylindrical quartz device that has an 8-cm internal diameter. Iron electrodes with dimensions of 11.5x6x0.2 cm were used as an anode and cathode. The inter-electrode distance was 3.5 cm and the effective surface area was 63.2 cm<sup>2</sup>. For each set, 250 mL of pre-coagulated leachate was used. Four UV lamps with 8 W power each were utilized for photo-oxidation. Around the outer surface of the quartz reactor, the lights are spaced equally apart. There was 5 cm of space left between the bulbs and the reactor. Figure 1 displays the schematic depiction of the setting of the experiment. Leachate's pH was changed by 6 N H<sub>2</sub>SO<sub>4</sub> and NaOH. First, experiments were implemented at different pH values by keeping other parameters constant to determine the pH effect in the UV/Electro-PS process. The effect of applied current, PS dose, and reaction time on COD,  $UV_{254}$ , and color removal efficiencies from pre-coagulated leachate were ascertained by implementing studies at 3-11 pH, 1-5 A applied current, 1-9 mM PS dose, and 5-35 min reaction time, respectively. 20 samples were used in the UV/Electro-PS process.

## RESULTS AND DISCUSSIONS

#### Effect of Process Parameters

UV/Electro-PS process was applied to the pre-coagulated leachate and the effect of the process parameters on the treatment efficiency was determined. Binary

activation processes of persulfate are more effective than single ones. The reason for this is the activation of persulfate by electrolytic or UV application and the formation of sulfate radicals with high oxidation potential. In the UV/Electro-PS process, the regeneration of Fe<sup>2+</sup> ions dissolved from the iron electrodes increases with the application of UV. Photoreduction of Fe(OH)<sup>2+</sup> to the dominant Fe<sup>3+</sup> species has a positive effect on hydroxyl radical formation (Equations 1-2) [26,27].



The impact of pH on COD,  $UV_{254}$ , and color removal efficiencies is given in Figure 2. The pH has a considerable influence on the electro-PS procedure using iron electrodes. An acidic pH level resulted in a greater rate of elimination. In advanced oxidation processes where persulfate is the oxidant, sulfate radicals are prevalent in acidic environments and neutral pH values, whereas hydroxyl radicals are dominant in alkaline settings, and near pH 9, both sulfate and hydroxyl radicals appear in approximately equal quantities in the solution [28–30]. At acidic pH levels, sulfate radicals predominate over hydroxyl radicals as radical species with a greater capacity for oxidation [31,32]. Given that the procedure uses iron electrodes, depending on the reaction time and current density, the occurrence of Fe<sup>2+</sup> ions, which get dissolved in the medium and function in the process as a coagulant and catalyst, rises as stated in Faraday's law. In the electro-PS set-up with iron electrodes, contaminant removal

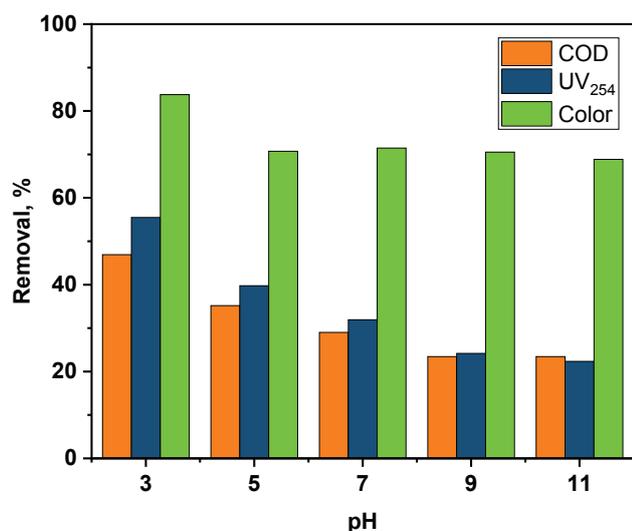
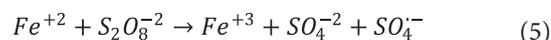
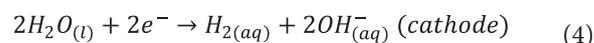
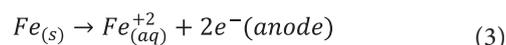


Figure 2. The effect of pH on the UV/Electro-PS process.

occurs through two mechanisms: coagulation ( $\text{Fe}(\text{OH})_n$ ) and oxidation ( $\text{SO}_4^{\cdot-}$ ). The pH plays a critical role in both of them. Coagulation including iron as a coagulant, is effective in the pH range of 4–9. At the same time, sulfate radicals are efficient in an acidic medium and interact with hydroxyl ions under alkaline conditions to form hydroxyl radicals. The efficacy of removing pollutants is reduced because hydroxyl radicals have a lower ability to oxidize than sulfate radicals [22,33,34]. It can be seen from Figure 2 that as the pH increases, there is a slight decrease in COD, UV<sub>254</sub>, and color removal efficiencies. The highest elimination yields were achieved at pH 3 and subsequent experiments were conducted at pH 3. The optimum pH value obtained in this study is in agreement with the previous studies which investigated electro-per-sulfate applications using different electrodes for the treatment of leachate or leachate nanofiltration concentrate [35–39]. The optimum pH for maximum pollutant removal is acidic both in this study and in the previous studies. When the current 2 A was applied and 5 g/L PS was added, COD, UV<sub>254</sub>, and color removal efficiencies were obtained as 47%, 55%, and 84%, respectively, at pH 3 in 15 minutes of reaction time.

The impact of applied current on the UV/Electro-PS process is presented in Figure 3. The experimental tests' findings demonstrated that the efficiency of removal rises with applied current. Sulfate radicals are produced in electrochemical procedures when persulfate is electrolytically activated [40,41].  $\text{Fe}^{2+}$  ions release off the iron plates and combine with the medium throughout electrolysis to activate persulfate electrolytically.  $\text{Fe}^{2+}$  ions ignite the persulfate in the solution, forming sulfate radicals and facilitating the elimination of pollutants. Equations 3–5 explain persulfate electrolytic stimulation [12].



Sulfate and hydroxyl radical production rates as well as quantity both rise with increasing applied current [42–45]. The effectiveness of removal rises as a consequence of a rise in radical production and quantity. The following equations describe how sulfate and hydroxyl radicals interact with organic materials.



Figure 3 shows that as the applied current increases, the removal efficiencies increase. The highest removal efficiencies were achieved when 5 A was applied; however, considering the cost and energy consumption, the optimum current was chosen as 3 A and the study was continued by applying 3 A. The optimum current values in electro-per-sulfate applications related to leachate treatment in the literature were reported between 1 and 2.1 A [35,37–39]. In this study, very high color removal efficiencies were obtained even at low current values; however, the optimum current value was chosen as 3 A to provide higher COD and UV<sub>254</sub> removal efficiency. The COD, UV<sub>254</sub>, and color removal efficiencies obtained at pH 3, 5 g/L persulfate dose, 3 A applied current, and 15-minute reaction time were 55%, 62%, and 88%, respectively.

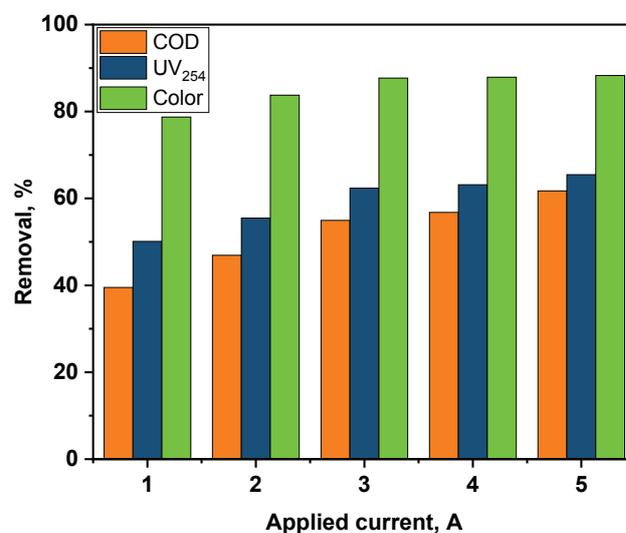
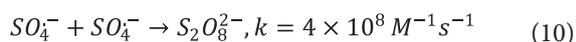
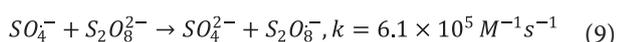


Figure 3. The effect of applied current on the UV/Electro-PS process.

One of the main elements influencing how well a treatment works is the dosage of persulfate [46]. Due to electrolytic stimulation, extra sulfate radicals will be produced if there is excess persulfate in the solution. Since these radicals possess a greater ability to oxidize than persulfate, the removal efficiency will be higher. Nonetheless, no consistency in the rise in efficiency of removal is contingent upon the persulfate dosage. This occurs due to the medium's restricted amount of substrate. Because there exists extra persulfate in the solution as the quantity of substrate drops, sulfate radicals interact with extra persulfate, forming sulfate ions and persulfate in the process (Equation 9). Simultaneously, persulfate is created when surplus sulfate radicals in the solution combine. Compared to sulfate radicals, persulfate has less oxidizing capability (Equations 8-10). The efficacy of removing organic debris is reduced as a result of these interactions, which deplete both sulfate radicals and persulfate.



In this study, experiments were implemented at 1-9 g/L persulfate doses to evaluate the effect of the persulfate dose, and it was observed that removal efficiencies up to 5 g/L dose increased and removal efficiencies decreased at higher doses (Figure 4). This situation can be explained by the formation of sulfate ions and persulfate as a result of the reaction of excess persulfate with sulfate radicals and sulfate radicals with each other. The optimum dose of persulfate

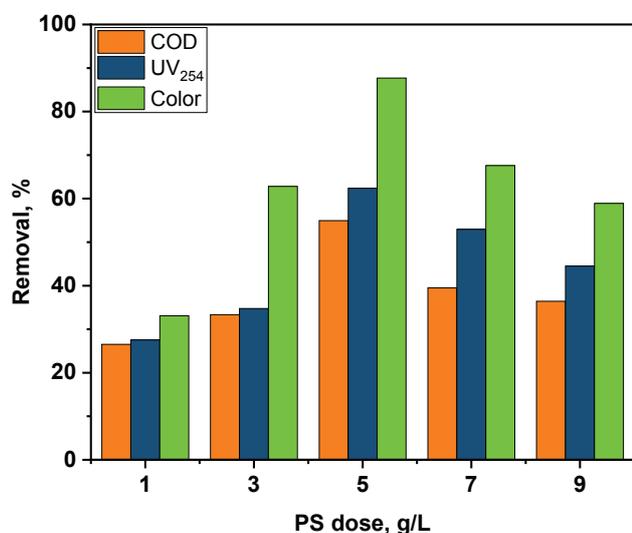


Figure 4. The effect of persulfate dose on the UV/Electro-PS process.

was chosen as 5 g/L. The initial COD value was 6424 mg/L and PS/COD ratio was determined as 0.77. Although this value is slightly lower than the PS/COD ratios obtained in previous studies, the obtained removal efficiencies are similar [35,37,38].

According to Faraday's Law, the reaction time precisely impacts persulfate activation [22]. It is observed that the removal yields rise as the reaction time rises (Figure 5). The longer the reaction time, the more  $Fe^{2+}$  ions dissociate from the electrodes into the medium and more persulfate is activated, which explains why elimination effectiveness rises with the length of the reaction. In the studies carried out to determine the effect of reaction time on pollutant removal, it was seen that as the reaction time rose, the pollutant removal yields rose. The optimum reaction time was selected as 25 min since a slight increase was seen at 35 min. In the previous studies, the optimum reaction time was reported as 34.8, 32.3, and 33.8 min, respectively by Varank et al. [37], Varank et al. [38], and Yazici Guvenc et al. [35]. The optimum reaction time in this study is lower than the previous studies even though the removal efficiencies are compatible. Under optimum conditions, (pH 3, applied current 3 A, 5 g/L persulfate dose, 25 min reaction time) COD, UV<sub>254</sub>, and color removal efficiencies were 55.6%, 68.5%, and 89.9%, respectively.

### Energy Consumption

The energy consumption due to both UV radiation and electricity consumption for the UV/Electro-PS process is calculated in kWh/kg COD using Equations 11-12 [47].

$$E_{uv} = \frac{P \times t \times 1000}{V \times \Delta COD} \quad (11)$$

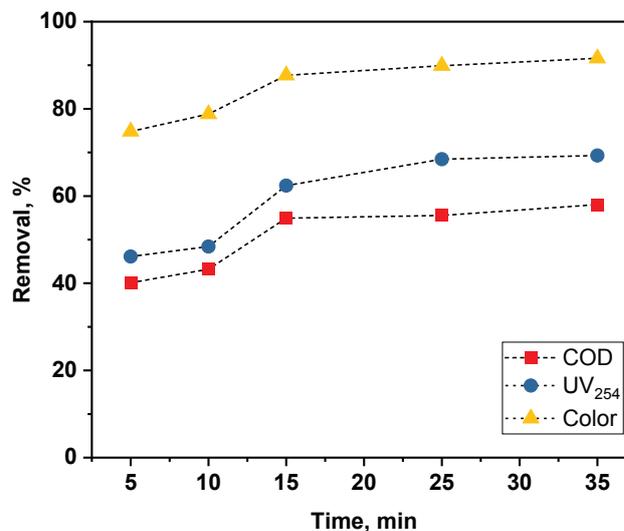


Figure 5. The effect of reaction time on the UV/Electro-PS process.

Where  $P$  is the power of the UV lamps (W),  $t$  is the illumination time of the lamps (h),  $V$  is the wastewater volume (L), and  $\Delta\text{COD}$  is  $\text{COD}_0 - \text{COD}_t$  (mg/L).

$$E_E = \frac{U \times i \times t}{V \times \Delta\text{COD} \times 1000} \quad (12)$$

Where  $U$  is the voltage (volts) applied to the wastewater,  $i$  is the electric current (A) supplied to the wastewater,  $t$  is the reaction time (h),  $V$  is the wastewater volume (L), and  $\Delta\text{COD}$  is  $\text{COD}_0 - \text{COD}_t$  (mg/L).

The energy consumed as a result of UV radiation was 17.0 kWh/kg COD and the consumed electrical energy was 10.4 kWh/kg COD and the total energy consumption was calculated as 27.4 kWh/kg COD.

## CONCLUSION

Ultraviolet and electrochemically activated persulfate (UV/Electro-PS) process was applied for COD, color, and  $\text{UV}_{254}$  removal from pre-coagulated landfill leachate. Optimum values of the pH, applied current, persulfate dose, and reaction time were determined for the UV/Electro-PS process. By applying the optimum conditions (pH 3, applied current 3 A, persulfate dose 5 g/L, 25 min reaction time) COD,  $\text{UV}_{254}$ , and color removal yields were 55.6%, 68.5%, and 89.9%, respectively. The total of the UV radiation and electrical energy consumption for the optimum process conditions was calculated as 27.4 kWh/kg COD. The study's findings demonstrated that UV/Electro-PS can be an effective post-treatment alternative for COD,  $\text{UV}_{254}$ , and color elimination from pre-coagulated landfill leachate. The results obtained in the study will create a base and shed light on the combined process applications in leachate treatment. Alternative studies are suggested that will reveal the effects of different coagulants and flocculants used in pretreatment in combined process applications and the effects of the different anode or oxidant use on process performance in UV/Electro-PS process applications.

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## AUTHORSHIP CONTRIBUTIONS

Authors equally contributed to this work.

## DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

## CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

## ETHICS

There are no ethical issues with the publication of this manuscript.

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