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Evaluating the effectiveness of advanced oxidation processes for leachate treatment: A systematic review

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Abstract

Background: Leachate, containing challenging-to-degrade organic substances and persistent toxins, poses significant environmental concerns. Advanced oxidation processes (AOPs) have emerged as a promising solution for effective leachate treatment. This research provides a comprehensive review of the impact of various AOPs in leachate treatment.

Methods: This systematic review was conducted, encompassing commonly used AOPs such as ozone, peroxone, O_3/catalyst, Fenton, photo-Fenton, UV/TiO_2, photolytic persulfate, O_3/UV, and O_3/H_2O_2/UV. Extensive searches were performed using reputable databases, including EBSCO, PubMed, Web of Science, and Google Scholar. Specific keywords and inclusion/exclusion criteria were applied. Data regarding leachate treatment parameters were meticulously summarized and analyzed using descriptive statistical methods.

Results: The efficiency of AOPs in removing leachate organic matter varied, with chemical oxygen demand (COD) removal ranging from 41% to 83% in treatment systems. The order of effectiveness was found to be: O_3/UV/H_2O_2 > photo-Fenton > UV/TiO_2 > Fenton > persulfate (PS) > O_3/UV > O_3/H_2O_2 > O_3/catalyst > ozonation (O_3). The highest COD removal efficiency of 83.75% was achieved using the O_3/UV/H_2O_2 AOP approach. The removal efficiency of color also varied, ranging from 32% to 100%, depending on the leachate's characteristics, concentration, and specific treatment process utilized.

Conclusion: AOPs, particularly the hybrid approach using O_3/UV/H_2O_2, significantly enhance waste leachate treatment by effectively degrading persistent organic compounds through the generation of hydroxyl radicals. Further research is required to optimize AOPs and improve their efficiency in waste leachate treatment.

Keywords: Advanced oxidation processes, Chemical oxygen demand, Hydroxyl radicals, Leachate treatment, Organic compounds

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Introduction

Effective management of leachate is a significant challenge in waste disposal, particularly in landfill operations. The interaction of physical, biological, and chemical processes within the landfill waste, combined with water infiltration and the initial moisture content of the waste, leads to the formation of a concentrated dark liquid known as landfill leachate (1). Landfill leachate, characterized by its high concentrations of harmful organic waste, poses a significant threat to the pollution of surface and subsurface water sources. Consequently, there is a pressing need to develop an effective method to enhance the efficiency of waste leachate treatment (2,3). The selection of an efficient method for treating solid waste leachate remains a significant challenge, as no universal solution has been identified (4,5). Conventional approaches to leachate treatment often have limitations and fail to completely mitigate the adverse environmental impacts associated with leachate (6). A general classification of leachate treatment technologies includes biological methods and chemical and physical methods, but due to the need to comply with strict quality standards for the direct discharge of leachate to surface water, the development of integrated treatment methods is required (5). Chemical

oxidation is required to treat wastewaters containing soluble organics that cannot be removed by physical separation and are non-biodegradable or for toxic biological oxidation (6). Advanced oxidation processes (AOPs) are an effective treatment method based on the production of hydroxyl radicals that oxidize stable organic matter and degrade pollutants in solid waste leachate, turning them into harmless products (7-9). AOPs can rapidly degrade diverse pollutants and significantly improve the biodegradability of the leachate (10). The use of advanced oxidation methods in wastewater treatment shows progress in the use of treatment methods that cause the breakdown of degradable and non-degradable hard materials into biodegradable compounds (11).

Various AOPs including ozone (O_3), ozone with hydrogen peroxide (O_3/H_2O_2), ozone with ultraviolet light (O_3/UV), hydrogen peroxide with ultraviolet light (H_2O_2/UV), Fenton process (H_2O_2/Fe^{2+}), and photo-Fenton process (H_2O_2/Fe^{2+}/UV) are commonly investigated for leachate treatment (12). Additionally, some studies have explored the combination of advanced chemical oxidation processes, such as ozonation along with the Fenton process ($O_3/H_2O_2/Fe^{2+}$), for wastewater treatment (13). These AOPs utilized for leachate and wastewater treatment can be categorized based on the physical and chemical mechanisms involved, as depicted in Figure S1 (see Supplementary file 1).

Table S1 presents a classification of advanced chemical oxidation processes utilized in leachate treatment, which includes non-photochemical and photochemical methods. Due to the low average biodegradability ratio of leachate, which is typically less than 0.1, the pollutant composition is too strong for biological processes. Therefore, advanced chemical oxidation is a suitable and effective treatment method for leachate (14,15). It is worth noting that AOPs are a subset of chemical processes.

Despite numerous studies exploring the effectiveness of different chemical oxidation processes in treating leachate, determining the most efficient and optimal treatment method continues to be a major challenge. This study aimed to investigate the effectiveness of different chemical oxidation processes in leachate treatment, highlighting the need for comprehensive comparative studies. This will help identify the advantages, limitations, and applicability of each method under different conditions. Systematic studies are crucial for comparative analysis, evidence-based decision-making, optimization of treatment processes, and sustainability advancements. The study emphasizes the importance of choosing effective and efficient methods due to the leachate's complex nature.

Materials and Methods

This study focused on a systematic review of research conducted in the field of using advanced chemical processes in leachate treatment to clearly understand the impact of each of those methods. The stages of review and data analysis included identifying articles, screening articles, determining the eligibility of articles, and then, finalizing articles that meet the inclusion criteria. No human subjects were involved in this study. Searches were performed using Library Services and databases of Mazandaran University of Medical Sciences (EBSCO, PubMed, etc.) and Google Scholar. This study employed a comprehensive approach to select relevant articles using keywords and inclusion/exclusion criteria. The keywords used included but were not limited to AOPs, leachate treatment, photocatalysis, waste management, and treatment outcomes. The Boolean search method was used to combine keywords to be more relevant to the topics of interest. Most of the reviewed sources included publications that were published up to 2023. One of the limitations of this study was not including all languages of the world because many articles were written in other languages (other than English).

The PICO model, which stands for Patient/Population, Intervention, Comparison, and Outcome, was utilized as a search strategy tool to guide the systematic study of leachate treatment. Specifically, the PICO model was employed to define the scope of the study by identifying the target population (leachate treatment), the intervention of interest (advanced chemical oxidation processes or AOPs), the comparison group (different AOPs), and the desired outcome (identification of the most effective AOP in leachate treatment). This structured approach ensured a focused and systematic search, as opposed to using another conceptualizing tool or conducting an unguided search.

Inclusion criteria included publications and sources with information regarding advanced chemical oxidation methods (AOP) in leachate treatment, as well as all other similar terms (e.g., oxidation, leachate, photocatalysis), the efficiency of AOP in removing pollutants from leachate and related physicochemical parameters and English language articles. Exclusion criteria included water and wastewater treatment, as well as other similar terms (e.g., coagulation/flocculation, air conditioner), non-scholarly articles and publications, and non-English language articles. The quality assessment and validation of the selected articles were done using the PRISMA checklist by two authors of this article separately.

A comprehensive search was conducted, resulting in the retrieval of a total of 74 articles for this research. These articles were then carefully reviewed, and a rigorous selection process was followed. Ultimately, 47 articles were deemed relevant and included in the study, as illustrated in Figure S2.

Among the included articles, a significant proportion (40 out of 47) were sourced from reputable databases such as PubMed and EBSCO. These databases are widely recognized for their focus on scientific literature

and scholarly research. The remaining 34 articles were sourced from various other platforms, including Google Scholar and other relevant sources.

This distribution of articles across different databases reflects a balanced approach to exploring the available literature on AOPs and leachate treatment. By incorporating multiple databases, the study aimed to ensure comprehensive coverage of relevant research and minimize potential bias associated with relying on a single source.

Statistical analysis

The data analysis process involved identifying information related to the effectiveness of advanced chemical oxidation in leachate treatment, as well as the efficiency of each relevant process. Similar studies were grouped and classified into relevant subgroups. The analysis focused on the efficiency of removing pollutants from the leachate, as well as the advantages and disadvantages of different chemical oxidation methods. The most pertinent information related to the subjects was identified, evaluated, analyzed, and grouped accordingly. Descriptive statistical methods were used to summarize and analyze the data about each of the leachate treatment parameters in the desired treatment methods.

Results

The objective of this study was to conduct a comprehensive analysis of AOP in leachate treatment. Through a systematic review of the literature, a significant amount of information related to AOP in leachate treatment was gathered. Various AOPs in leachate treatment were identified and examined in this literature review.

Overview

In this study, a total of 2800 articles were searched and 47 full-text articles were investigated. These studies focused on the treatment of landfill leachates using various AOPs. Many of the studies employed combined methods with AOPs for leachate treatment. All the research studies included in this analysis were original and specifically studied leachate treatment. The study categorized different methods for integrating leachate treatment into three parts: non-photochemical methods (O_3/H_2O_2, O_3, O_3/catalyst, Fenton), photochemical methods (O_3/UV, O_3/UV/H_2O_2, photo-Fenton, and photo-Fenton like UV/TiO_2), and persulfate (PS) oxidation (different processes of PS oxidation).

Different ways to integrate leachate treatment

This study examined 9 common AOPs for waste leachate treatment that have been used in various studies. Many of the studies have employed combined methods for optimal and complete leachate treatment. The choice of the process type depends on the physical and chemical characteristics of the leachate and other factors. To facilitate understanding, the processes were classified into two groups: photochemical and non-photochemical groups.

Non-photochemical methods (generating hydroxyl radicals without light energy)

In the non-photochemical methods, the production of hydroxyl radicals occurs without the need for light energy. In this part of the research, relevant articles in the field of leachate treatment using non-photochemical methods were searched and analyzed. The investigation focused on four separate processes including ozonation, peroxone, ozone/catalyst, and Fenton.

Ozonation

Outcomes and efficiencies related to the use of ozone (O_3) processes in landfill leachate treatment include significant •OH production, reduced COD and increased BOD_5/COD ratio, the biodegradability of leachate, and color removal, which were identified in this systematic literature review (Table 1). Chemical oxidation processes are capable of treating strong pollutants such as landfill leachates and are considered strong oxidation processes. Considering its strong oxidizing agent, AOPs using O_3 have been widely used by researchers in leachate treatment. Hydroxyl radical (•OH) is a strong and non-selective chemical oxidant that reacts very quickly with most organic matter compounds (16,17). Ozone is a synthetic environmental agent with no harmful residues (18). The use of O_3 in combination with other treatment processes improves the biodegradability of hard degradable organic materials (19). The results of recent studies have shown that ozonation plays an important role in the advanced treatment of highly resistant wastewater such as biologically treated leachate variability caused significant variation in the O_3 utilization efficiency (O_3E) for COD removal (20). The oxidation process is usually combined with a subsequent process (e.g., adsorption and biological treatment) or a recycling stream, which can reduce operating costs and achieve high-quality final effluent (21,22). Treatment outcomes appeared to include the removal efficiency of COD (41.4%) and color (93.0%).

Ozone and hydrogen peroxide (peroxone)

There were different results and efficiencies related to the use of O_3 and H_2O_2 processes in landfill leachate treatment, which was identified in this systematic literature review (Table 1). Outcomes and efficiencies related to the use of O_3 and H_2O_2 processes in landfill leachate treatment include improvement of biodegradability (BOD_5/COD ratio up to 0.36), the increased removal efficiency of COD and total organic carbon (TOC), the biodegradability of leachate, the highest synergistic effect of O_3 with H_2O_2, and color removal. Ozone combined with H_2O_2 is an efficient

Table 1. Comparison of treatment of landfill leachate under different ozonation oxidation processes (O_3, O_3/H_2O_2, O_3/catalyst)

AOP features	Landfill leachate characteristics	Removal efficiency (%)	Conclusion	Source
O_3 dose = 0.48 mg O_3/ mg COD	COD = 706-1846 mg/L BOD_5 = 20-50 mg/L pH = 8-8.5	COD = 33%	Ozonation increased the adsorption capacity and breakthrough time of GAC by a factor of 2.5 compared to flocculation-coagulation.	(23)
O_3 dosage = 159 mg/L Gas flow rate = 0.5 LPM Time = 50 minutes pH = 11	COD = 39333.33 mg/L BOD = 284.4 mg/L pH = 8.57	COD = 46.4%	Ozonation alone is effective in leachate treatment.	(24)
O_3 dosage = 27 g/N m^3 pH = 8.2 Reaction time = 60 min	BOD_5 = 40–60 mg/L COD = 2100–2580 mg/L NH_4-N = 1000–1160 mg/L pH = 8.2-8.4	Color = 93% COD = 80% NH_4-N = 71%	A combined treatment resulted in the removal of inorganic and small non-degradable organic molecules, which reduced COD and increased the BOD_5/COD ratio.	(17)
Time interval. Qg (l/min) = 0.83 l/min O_3 concentration = 112 mg/L pH = 7	COD = 743 mg/L TOC = 284 mg/L	COD = 30% TOC = 21%	After ozonation, BOD_5 increased about 80% due to the increase in the degradability of leachate materials.	(25)
Time interval. Qg (Lmin) = 1 L/min O_3 concentration = 80-90 mg/L pH = 10	Biologically treated pH:8.2-8.5 COD = 1392 mg/L	COD = 16%	Ozonation of landfill leachate led to a significant •OH production.	(26)
O_3 = 40 L/h O_3 doses = 0.28 to 1.4 g/dm^3 COD/H_2O_2:1/2 pH = 3	TOC = 51 mg/dm^3 BOD_5 = 81 mg O_2/dm^3 COD = 267 mg O_2/dm^3	TOC = 33.4% BOD_5 = 75% COD = 79%	Improvement of biodegradability (BOD_5/COD ratio up to 0.36).	(27)
H_2O_2 = 600 mg/L pH = 9 Time = 60 min Qg = 0.83 L/min CO3,I-g = 112 mg/L	COD = 743 mg/L TOC = 284 mg/L pH = 3.5	COD = 63% TOC = 53%	The combined effect of ozone with H_2O_2 increased the removal efficiency of COD and TOC, as well as the biodegradability of leachate.	(25)
H_2O_2 = 400 mg/L pH = 9 Time = 180 min Qg = 0.5 L/min H_2O_2 = 4 g/L	COD = 1880 mg/L pH = 7.1	COD = 43%	Ozone combined with H_2O_2 are efficient methods to treat this leachate.	(28)
H_2O_2 = 500 mg/L Time = 180 min pH = 9 O_3 dose = 18 mg O_3/min	pH = 3.7 DOC = 395 mg/L COD = 1073 mg/L	COD = 57% DOC = 49%	The highest synergistic effect was obtained.	(29)
H_2O_2 = 2 g/L O_3 concentration = 80 g/m^3 NTP ± 0.5% Gas flow rate = 200 mL/min ± 10% pH = 8.7	COD = 5230 mg/L BOD_5 = 500 mg	COD = 48% Color = 94%	Ozone combined with H_2O_2 is an efficient method to treat this leachate.	(30)
g/L(Nano-Fe_3O_4@CDA) + O_3 Catalyst dosage = 1.0 O_3 dosage = 3.0 g/L Reaction time = 120 min pH = 7	COD = 1050 mg/L BOD_5/COD = 0.05 pH = 8.48	COD = 53%	Catalytic ozonation improved the removal of hazardous organics, increased the biodegradability of the leachate, and made the subsequent MBR process favorable.	(31)
Ozone/manganese (M + O_3) O_3 dosage = 2.882 g/h Manganese ore = 300 mg/L Time = 60 minutes pH = 8	COD = 3,083 mg L^{-1} TOC = 980 mg L^{-1} Color = 2,536 Pt-Co	COD = 20% TOC = 14%	The amount of manganese ore greatly affected the removal of organic compounds from the landfill leachate after the pH parameter.	(24)
Ozone/ZrCl4 (COD/ZrCl4 ratio) = 1 g/1 Ozonation time = 60 min O_3 consumption = (3.81 kg O_3/kg COD) pH = 8	BOD5 = 60 mg/L COD = 1123 mg/L (BOD_5/COD) = 0.05-0.11 NH_4-N = 620 mg/L pH = 8.10 Color = 2982 PtCo	COD = 48%, Color = 75%, NH_3-N = 69%	The oxidation performance of $O_3/ZrCl_4$ is efficient in the treatment of stabilized leachate.	(19)
Ozone/$ZrCl_4$ (COD/ZrCl4 ratio) = 1 g/1 g Ozonation time = 60 min O_3 consumption = (2.32 kg O_3/kg COD) pH = 8	BOD_5 = 227 mg/L COD = 2180 mg/L (BOD_5/COD) = 0.07-0.08 NH_4-N = 1897 mg/L pH = 7.93 Color = 4650 PtCo	COD = 33% Color = 70% NH_3-N = 53%	The oxidation performance of $O_3/ZrCl_4$ is efficient in the treatment of stabilized leachate.	(19)
Fe−NiCAF catalyst + O_3 Fe−$_{Ni}$CAF dosage = 4 g/L^{-1} Gas flow rate = 0.5 L/min^{-1} O_3 concentration = 20.0 mg/L^{-1}	COD = 1720 mg/L	COD = 57%	Fe−$_{Ni}$CAF catalyzed ozonation was highly efficient for landfill leachate treatment.	(32)

method to treat leachate. Treatment outcomes appeared to include the removal efficiency of COD (58.0%), TOC (43.2%), BOD_5 (75.0%), and color (94.0%). The primary can-be-changed AOP features discussed in the literature were O_3 doses and H_2O_2, time, and landfill leachate characteristics. Primary landfill leachate characteristics included TOC, BOD_5, pH, COD, and other leachate characteristics. The COD/H_2O_2 ratio and the O_3 gas flow rate were discussed in some literature on landfill leachate treatment under different O_3/H_2O_2 oxidation processes.

Ozone + catalyst (O_3/catalyst)

Catalytic ozonation improved the removal of hazardous organics, increased the biodegradability of the leachate, and made the subsequent process favorable. The catalysts used included Nano-Fe_3O_4@CDA, manganese, $ZrCl_4$, Fe–NiCAF, and other catalysts related to oxidation processes. The characteristics of the landfill leachate and the catalyst's characteristics were investigated in the studies (Table 1). Most of the catalytic materials in a column reactor with O_3 or peroxone system and with increased contact surface improved landfill leachate treatment. The amount of manganese ore greatly affected the removal of organic compounds from the landfill leachate after the pH parameter. Fe–NiCAF catalyzed ozonation was highly efficient for landfill leachate treatment. The oxidation performance of O_3/$ZrCl_4$ is efficient in the treatment of stabilized leachate. Treatment outcomes appeared to include the removal efficiency of COD (42.2), NH_3-N (61.0%), and color (72.5%). The removal efficiency of TOC was lower compared to other parameters. Catalyst type, catalyst dose, pH, reaction time, and leachate characteristics were among the most important parameters discussed in different studies.

Fenton process (H_2O_2/Fe_2+)

Most of the reviewed articles recommend different methods of leachate treatment combined with AOP (42 articles). From all the articles reviewed in the field of waste leachate treatment under different Fenton oxidation processes, five studies whose full text was available and all the parameters required in this study including leachate characteristics, AOP characteristics, removal efficiency, findings, and conclusions were reviewed (Table 2). H_2O_2 and Fenton's treatment increases the leachate treatment efficiency by increasing the COD removal rate. Fenton oxidation increased the biodegradability of the leachate and removed the refractory compounds (non-biodegradable organic matter) of the leachate effluent. The electro-Fenton method used with the help of chemical coagulation was very efficient in removing a large amount of refractory organic and inorganic compounds in leachate. It seems that the results of leachate treatment with the Fenton process include removal efficiency of COD (69.26%), NH_3-N (74.2%), and color (>98%). Fenton oxidation was successful in removing the aromatic structure and C=C and C=O bonds in MCLL. According to Table 2, the comparison of landfill leachate treatment under different Fenton oxidation processes showed that the effect of AOP characteristics and landfill leachate characteristics are very important in the removal of leachate pollutants. Iron concentration is effective. Also, the results show the optimal efficiency of the Fenton process in removing leachate pollutants at acidic pH (pH<4) and this is one of the characteristics of the Fenton process. The reaction time in the Fenton process was from 30 to 60 minutes in different studies.

Photochemical methods

According to the division of AOPs based on two main processes, in this part of the article, common photochemical processes were reviewed in five sections. Photochemical processes involve irradiation to achieve complete oxidation and oxidative degradation, which makes compounds resistant to oxidation. They become more sensitive to unaided oxidants (36). By reviewing related articles, all types of photochemical processes and their characteristics, as well as the efficiency of removing organic matter in leachate treatment, were investigated and analyzed in the studies.

O_3/UV

O_3/UV significantly increased the biodegradability of the leachate. Acute toxicity of the leachate significantly decreased after VUV/O_3 treatment. UV plays an effective role in the O_3/UV oxidation process in leachate treatment, especially in concentrations of ion and turbidity. The combined process of O_3/UV seems to be more effective in removing organic substances, pollutants, and toxic substances from leachate at higher pHs. The results of the comparison of the treatment of landfill leachate under different O_3/UV oxidation processes are shown in Table 3. AOP characteristics including O_3 dose, pH, lamp power (W), and irradiation time were important parameters discussed in the reviewed studies. In some studies, H_2O_2 was also included in the process. COD, TOC, and PH were among the most important properties of treated leachate. Different studies show that the reaction time is in the range of 60 to 90 minutes. Treatment outcomes appeared to include the removal efficiency of COD (61.7%), TOC (43.5%), and color (73.3%). O_3/UV systems performed better than O_3 alone for COD and color removal. In addition, O_3/UV oxidation processes can effectively change the distribution of dissolved organic matter and fluorescence characteristics of concentrated leachate.

H_2O_2/UV and O_3/H_2O_2/UV

In this part of the study, the results of the H_2O_2/UV and O_3/H_2O_2/UV oxidation processes are presented (Table 3). As shown in this table, the synergistic effect of O_3 and H_2O_2

Table 2. Comparison of treatment of landfill leachate under different Fenton oxidation processes

AOP Features	Landfill leachate characteristics	Removal efficiency (%)	Conclusion	Source
(H_2O_2/Fe^{2+}) H_2O_2 = 66.47 mμ/L Time = 60 minutes Fe^{2+} = 80 mμ/L pH = 3	COD = 39333.33 mg L^{-1} BOD = 284.4 mg L^{-1} pH = 3	COD = 52.91%	H_2O_2 and Fenton's treatment increases the leachate treatment efficiency by increasing the COD removal rate.	(24)
(O_3/Fe^{2+}) O_3 dosage = 159 mg/L Gas flow rate = 0.5 LPM Time = 75 minutes Fe^{2+} = 80 mμ/L pH = 3	COD = 39333.33 mg L^{-1} BOD = 284.4 mg L^{-1} Nitrogen = 375.2 mg L^{-1}	COD = 80%	Compared to the ozonation and Fenton process alone, COD reduction performance improved by 30%.	(24)
(H_2O_2/Fe^{2+}) $FeSO_4 \cdot 7H_2O$ = 20 gl^{-1} H_2O_2 = 20 ml l^{-1} pH = 3	BOD_5 = 619.3 mg/L^{-1} COD = 1625 mg/L^{-1} NH_3–N = 30 mg/L^{-1}	BOD = 15.3% COD = 60.8% NH_3-N = 67.4%	Fenton oxidation removed the refractory compounds (non-biodegradable organic matter) of the leachate effluent.	(1)
(Fe_2 + /H_2O_2) H_2O_2 = 15000 mg/L Catalyst = 2000 mg/L	COD = 3823 mg/L^{-1} TOC = 2120 mg/L^{-1} BOD = 680 mg/L^{-1} PH = 7.94	COD = 58%	Iron concentration corresponding to a Fe^{2+}/COD mass Ratio = 0.33 was found to be the most favorable.	(33)
Coagulation + electro-Fenton + SBR H_2O_2 = 750 mg/L Time = 30 minutes pH = 4	COD = 1941 mg/L^{-1} NH_3–N = 150.9 mg/L^{-1}	COD = 85% NH_3-N = 81% Color = 100%,	The electro-Fenton method with the help of chemical coagulation was very efficient in removing a large amount of refractory organic and inorganic compounds in leachate.	(34)
H_2O_2/Fe (II) H_2O_2 = 9.0 mL/200 ML H_2O_2/Fe (II) molar ratio = 3.0 Time = 40 minutes pH = 3	Leachate concentrate after nanofiltration	COD = 78.9 ± 1.3% TOC = 70.2 ± 1.4%	The aromatic/C = C structure and C = O bonds in MCLL were successfully removed by Fenton oxidation.	(35)

is effective in leachate treatment, and the tested ozone-based AOP processes (O_3/H_2O_2/UVC) lead to the highest synergistic effect and increased biodegradability. The investigation of landfill leachate treatment under different O_3/UV/H_2O_2 oxidation processes showed that this process is an effective pretreatment or treatment technology for landfill leachate. In the UV/H_2O_2 oxidation test, the removal efficiency of TOC and color increased with increasing H_2O_2 dosage. Treatment outcomes appeared to include the removal efficiency of COD (75.12%), and color (83.75%). In the UV/H_2O_2 process, with the increase of H_2O_2 dosage, the removal efficiency COD and color of the leachate increased and better performance was obtained than the H_2O_2 system alone in leachate treatment. The characteristics of raw landfill leachate are shown in Table 3. A low BOD_5/COD ratio indicates low biodegradability of leachate. For all pH values (3, 4, 6, and 9), COD, TOC, TKN, and color removal were observed. For pH 3 and 4, COD removal was higher.

Photo-Fenton and photo-Fenton like

UV-Fenton (combination of H_2O_2 and ferric iron with ultraviolet radiation) is an AOP that has been shown in various studies to be a very effective and suitable technology for the treatment of concentrated leachate with a high content of organic matter (45-47). In the Fenton process, H_2O_2 is used as an oxidizing agent, and the reduced form of iron (Fe^{2+}) is used as a catalyst to create the $^{\bullet}OH$ point under acidic conditions (48,49). The efficiency of the Fenton photo-oxidation process is higher due to the higher production of hydroxyl radicals compared to the Fenton process. Photo-Fenton > Fenton-like > Fenton processes are effective technologies to reduce COD in leachate. Photo-Fenton process was the most effective treatment to reduce COD in landfill leachates. The comparison of landfill leachate treatment under different photo-Fenton oxidation processes is shown in Table 4. Copper (Cu^{2+}) is used in the Fenton-like process. Fe^{2+} doses of 500 to 2000 mg L^{-1} have been reported in various studies. The properties of leachate and new AOP are decisive in catalyst dosage, reaction time, and UV intensity. Treatment outcomes were the removal efficiency of COD (81.25%) and color (99.97%).

Photocatalysis (UV/TiO_2)

Catalysts and light with sufficient intensity are necessary to carry out photocatalytic processes (52,53). The effect of each of the parameters (UV and TiO_2) alone in removing the organic load of leachate is insignificant. However, their synergistic effect increases the purification efficiency/ TiO_2 photocatalysis is very effective in the decolorization and mineralization of landfill leachate. Landfill leachate characteristics, AOP features, and removal efficiency (%) were investigated in different studies (Table 5). Treatment outcomes appeared to include the removal efficiency of COD (70.60%) and color (66.3%). Nanoparticle dose, reaction time, and pH were among the most important parameters discussed in the studies. After UV/TiO_2 photocatalytic treatment, landfill leachate becomes biodegradable and can be treated by secondary biological

Table 3. Comparison of treatment of landfill leachate under different oxidation processes (O_3/UV, O_3/UV/H_2O_2)

AOP features	Landfill leachate characteristics	Removal efficiency (%)	Conclusion	Source
Dissolved O_3 loading = 3.4 mg O_3 per g COD Time = 90 min H_2O_2 concentration = 2.82 g L^{-1}	Fluorescence matters = 185.6 mg L^{-1}	Fluorescence matters = 75.1% COD = 41.7% TOC = 8% Color = 35%	In O_3-combined processes, the removal efficiencies of COD, TOC, and color increased by approximately 10-15%, 7-15%, and 15-20%, respectively.	(37)
pH = 11.86 O_3 dosage = 50 mg/min VUV lamp = 14 W Time = 60 minutes	COD = 740-800 mg/L TOC = 302.4 mg/L	COD = 88% TOC = 61.75%	Incineration leachate membrane concentrate (ILMC) was effectively treated by VUV/O_3.	(38)
O_3 dose = 2.1 g O_3/L UVC = 12.2 kj_{uvc}/L	DOC = 956 mg/L^{-1} COD = 3479 mg/L^{-1} PH = 7.5	DOC = 73.2% COD = 79.4% Color = 97.8%	O_3/UVC significantly increased the biodegradability of leachate (more than 70%).	(39)
O_3 dosage = 11.2 mg min^{-1} Time = 60 minutes pH = 9	COD = 390 mg/L^{-1} BOD_5 = 50-70 mg/L^{-1} pH = 9	COD = 36.4% BOD_5/COD = 0.73	Acute toxicity of the leachate significantly decreased after VUV/O_3 treatment.	(40)
O_3 = 50 mg/min^{-1} PH = 7.8	BOD = 2920 COD = 26000	TOC = 61% COD = 63% Color = 87.2%	UV plays an effective role in the O_3/UV oxidation process in leachate treatment, especially at low concentrations and turbidity.	(41)
H_2O_2/UV H_2O_2 = 26g/L UV = 1500W pH = 3	COD = 14600 BOD_5 = 2920	COD = 96% BOD/COD = 0.45 TOC = 78.9% Color = 95.5%	In the UV/H_2O_2 oxidation test, the removal efficiency of TOC and color increased with increasing H_2O_2 dosage.	(41)
H_2O_2/UV H_2O_2 dose = 3 g/dm^3 Radiation time = 1.5 h PH = 4	COD = 1900 to 2700 mg O_2/dm^3 BOD_5/COD = 0.16	COD = 74.6%	Using the process (H_2O_2/UV) in which strong oxidative agents (hydroxyl radicals) are produced was effective for leachate treatment.	(42)
H_2O_2/UV H_2O_2 concentration = 232.7 mM T = 300 min UV = 4-UV lamps H_2O_2 = (35%, w/w)	TS = 7750 mg/L SS = 1150 mg/L pH = 7.8 COD = 3750 mg/L PtCo color unit = 8250	COD = 65% Color = 72%	The UV/H_2O_2 process was an effective pretreatment or treatment technology for landfill leachate.	(43)
O_3/UV/H_2O_2 UVC = 6 W H_2O_2 = 500 mg/L pH = 9 T = 180 min *Qg* = 0.1 L/min $C_{O3,l\text{-}g}$ = 180 mg/L	pH = 3.7 DOC = 395 mg/L COD = 1073 mg/L	COD = 67% DOC = 58%	Ozone-based AOP processes tested (O_3/H_2O_2/UVC), leading to the highest synergistic effect and biodegradability enhancement.	(29)
O_3/UV/H_2O_2 O_3 = 120 L/h UV = 991 J/cm H_2O_2 concentration = 1 g/L pH = 6	BOD_5 = 513 mg/L TSS = 779 mg/L TKN = 335 mg/L pH = 8.9 COD = 10605 mg/L	COD = 73% TKN = 76% TSS = 79%	Electrochemical treatment followed by precipitation can effectively reduce leachate nutrients and color.	(44)

Table 4. Comparison of treatment of landfill leachate under different Photo-Fenton oxidation processes

AOP features	Landfill leachate characteristics	Removal efficiency (%)	Conclusion	Source
(Fe^{2+}/H_2O_2/UV) H_2O_2 = 2720 mg L^{-1} Fe^{2+} = 544 mg/L^{-1} UV = 25 W pH = 4	COD = 3332 mg/L^{-1} BOD = 141 mg L^{-1} PH = 8.3	COD = 75%	The Fenton process is an effective technology for reducing the COD in leachate.	(50)
(UV/ Fe^{2+}/H_2O_2) Catalyst = 2000 mg/L Fe^{2+} H_2O_2 = 10000 mg/L Time = 60 min	COD = 3823 mg/L^{-1} TOC = 2120 mg/L^{-1} BOD = 680 mg/L^{-1} PH = 7.94	COD = 86%	Photo-Fenton process was the most effective treatment to reduce COD in landfill leachates.	(47)
(UV/Cu^{2+}/H_2O_2) Catalyst = 1000 mg/L Cu H_2O_2 = 5000 mg/L Time = 60 min	COD = 3823 mg/L^{-1} TOC = 2120 mg/L^{-1} BOD = 680 mg/L^{-1} PH = 7.94	COD = 66%	Photo-Fenton > Fenton-like > Fenton	(47)
Fe^{2+}/H_2O_2/UV H_2O_2 dosage = 2720 mg L^{-1} Fe^{2+}dosage = 544 mg L^{-1} UV = 25 W	PH = 8.3 COD = 3332 mg/L BOD_5 = 141mg/L	COD = 75%	The Fenton process is an effective technology for reducing the COD in samples of this leachate.	(50)
Fe^{2+}/H_2O_2/UV SUVA= 100.UV-254 pH = 4.75 Time = 6 h	BOD_5 = 58 TOC = 1802 mg C L^{-1} pH = 8.30 COD = 4897 mg O_2 L^{-1}	Color = 99.97% COD = 89%	Biodegradability (BOD_5/COD) increased by over 0.3.	(51)

Table 5. Comparison of treatment of landfill leachate under different UV/TiO_2 oxidation processes

AOP features	Landfill leachate characteristics	Removal efficiency (%)	Conclusion	Source
TiO_2 = 2.0 g/L Time = 30 minutes UV = 15 W Temperature = 25 ± 1°C pH = 4	COD = 2440.3 mg L^{-1} BOD = 225.4 mg L^{-1} TOC = 913.8 pH = 8.24	COD = 60% DOC = 70% Color = 97%	UV/TiO_2 photocatalysis is very effective in the decolorization and mineralization of landfill leachate.	(54)
Nano-TiO_2 dosage = 2 g/L Time = 72 h pH = 5	TOC = 930 BOD_5 = 225 mg L^{-1} DOC = 914 pH = 8.24	COD = 60% DOC = 74%	After UV/TiO_2 photocatalytic treatment, landfill leachate becomes biodegradable in nature and can be treated by the secondary biological treatment.	(55)
TiO_2 = 1 g/L UV lamp (model GGZ-125, 125 W; k 365 nm) pH = 5	BOD_5 = 10000-20000 mg/L^{-1} COD = 5000-10000 mg/L^{-1} TOC = 9000-15000 mg/L^{-1} PH = 6.5-7.5	COD = 82%	Coupling ARB with AOPs is a potentially applicable process to deal with bio-recalcitrant compounds present in mature landfill leachate.	(56)
TiO_2 nanoparticles = 4 g/L Sunlight = 500 W Time = 30 min pH = 8.88	COD = 7647 mg/L^{-1} NH_4^+–N = 26.73 mg/L PH = 8.8	COD = 89% Color = 70%	TiO_2/UV photocatalyst can be a technology to accelerate the leachate treatment process.	(57)
TiO_2 = 60 g/m^2 Time = 20 h UV = 77 W pH = 5	COD = 450 mg/L^{-1} BOD_5 = 15 mg/L^{-1} PH = 9-10	COD = 62% Color = 32%,	The UV/TiO_2 photocatalytic process can reduce the organic load of leachate for use in agriculture.	(58)

treatment.

Persulfate advanced oxidation process

PS AOP is an advanced technology in the field of landfill leachate treatment. The application of PS in landfill leachate treatment is systematically reviewed in this paper (Table 6). In the treatment of landfill leachate, PS has significant oxidation ability, stability, and high solubility. Heterogeneous catalysts as well as heat, UV, MW, and microwave activation can all activate PS. Among the activators used in various studies, heterogeneous catalysts with the advantages of high efficiency, low energy consumption, and satisfactory stability have been of interest for further use and are more promising (59-61). The Fe (II) activation-PS process is effective for COD removal from leachate nanofiltration concentrate. Among the various compounds in the PS oxidation processes, the best treatment efficiency was achieved with the UV-PMS process. This process also consumed the least amount of electrical energy. Much less PS is typically dosed for the pretreatment of landfill leachate. It seems that the results of leachate treatment with PS oxidation process include COD (67.87%) and color (32.0%) removal efficiency. This process had a good efficiency in leachate treatment in the pH range of 3 to 11. Heat, UV, and Fe (II) were common activators. According to the characteristics of the raw leachate, the PS dose was up to 200 mg/L and the time of UV radiation was up to 250 minutes. In some processes, the synergistic effect of H_2O_2 is used. The results of the studies show that the temperature up to 60 degrees had a good effect on the optimal progress of the process.

Comparison types of AOPs in leachate treatment

Hydroxyl radicals produced in the AOP process have oxidation potentials in the range of 2.8 V to 1.95 V (pH = 0 to 14) against the standard reference electrode, which determines the efficiency of the process. The hydroxyl radicals function as highly reactive oxidizing agents in leachate treatment. Figure 1 illustrates the oxidation potential of various common and potent oxidizing radicals (67-69). As per Figure 1, the hydroxyl radical exhibits the second-highest oxidizing potential, following fluorine. Due to its non-selective oxidation power, the hydroxyl radical reacts rapidly with many species with a rate constant of in the range 10^8–10^{10} M^{-1} s^{-1} (70). Hydrogen peroxide and superoxide are more reactive species that are formed during subsequent reactions and can degrade and destroy resistant organic materials in leachate. Oxidizing agents, radiation, and catalysts must be combined to create OH in situ since it has an extremely limited lifespan (71).

The review of various studies in the field of waste leachate treatment with advanced chemical oxidation in this review shows that researchers have tested different methods of advanced chemical oxidation under different conditions and for leachates with different characteristics in different regions of the world. The performance of each of the different processes used has been different. The effort to achieve the best performance in this field continues. By comparing similar studies with close conditions, each of the processes O_3/UV/H_2O_2 > Photo-Fenton > UV/TiO_2 > Fenton > PS > O_3/UV > O_3/H_2O_2 > O_3/catalyst > ozonation (O_3) performed well in removing COD and improving effluent from leachate treatment (Figure 2). Some processes are more effective in removing and destroying some pollutants, so the percentage of removal parameters in leachate is different. The nature of the leachate and considering other conditions and cost-relevant factors are very important in choosing the composition of the treatment system. The effectiveness of AOP techniques depends on the degree of removal of resistant pollutants, cost-effectiveness, technical

Table 6. Comparison of treatment of landfill leachate under different persulfate oxidation processes

AOP features	Landfill leachate characteristics	Removal efficiency (%)	Conclusion	Source
$S_2O_8^{2-}$:12COD0 = 2 Temperature: 50°C pH = 4 $S_2O_8^{2-}$ (Thermal activation)	COD = 1254 mg/L^{-1} SO_4^{2-} = 0 mg/L pH = 8.3	COD = 91%	SR-AOP appears to be more advantageous over hydroxyl radical (•OH)-based advanced oxidation processes (HR-AOPs) because •OH almost does not oxidize ammonia.	(62)
$S_2O_8^{2-}$ = 0.2 g L^{-1} H_2O_2 = 0.67 g L^{-1} Irradiation = 250 min Zeolite for subsequent adsorption = 1 g L^{-1} UV solar/O_3/H_2O_2/$S_2O_8^{-2}$	COD = 6500 mg/L^{-1}	COD = 96% Color = 32%	In the UV solar/O_3/H_2O_2/$S_2O_8^{-2}$ process, the interaction of $S_2O_8^{-2}$ and •OH leads to the oxidation of more organic compounds.	(63)
$S_2O_8^{2-}$/COD = 5.2 Temperature = 80°C pH = 10.9 Time = 120 Min $S_2O_8^{2-}$ (Heat activation)	COD = 5575 mg/L^{-1} Color = 11850 Pt/Co pH = 7.8	COD = 93.5%	Heat-activated PS process may be an alternative technology for COD and inert COD removal from leachate nanofiltration concentrate.	(64)
$S_2O_8^{2-}$/COD = 6.7 Fe (II) = 90 mM pH = 3 $S_2O_8^{2-}$ (Fe (II)activation)	COD = 5575 mg/L^{-1} Color = 11850 Pt/Co pH = 7.8	COD = 76.2%	The Fe (II)activation -PS process is effective for COD removal from leachate nanofiltration concentrate.	(64)
$Na_2S_2O_8$, > 98% PH = 8.2 Temperature = 60°C SR-AOPs ($Na_2S_2O_8$)	COD = 1096 mg/L NH_3-N = 560 mg/L pH = 8.2	COD = 81%	Much less PS is typically dosed for the pretreatment of a landfill leachate.	(65)
Absorbance = 254 nm (UV254) 15.18 cm^{-1} Oxidant dosage (PMS) = 0.048 mol/L UV-PMS	pH = 7.86 NH^{4+}-N = 1456.76 mg/L COD = 5680 mg/L	COD = 37.39%	The best treatment efficiency was achieved by the UV-PMS process, and this process also consumed the least electrical energy.	(66)

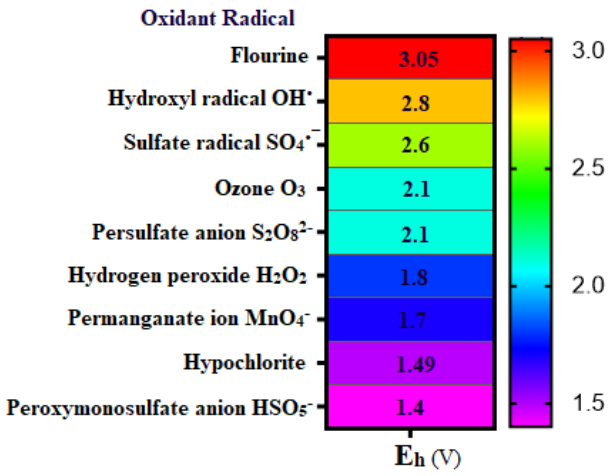

Figure 1. Oxidation potential for some common oxidants

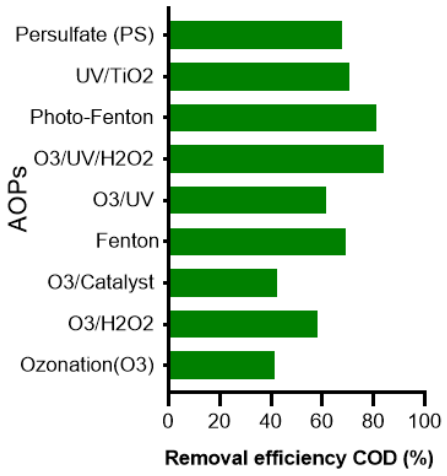

Figure 2. Comparison of AOP$_s$ in terms of COD removal efficiency

durability, and compatibility with the environment (72). Comparing the processes, 71-76% of COD was removed from old landfill leachate using Fenton oxidation (73), while 83% of COD was degraded using photo-Fenton (74). The performance evaluation of different AOPs in landfill leachate treatment and the findings of many researchers show that AOPs are efficient in reducing COD and improving biodegradability for both young and stabilized landfill leachate. Ozone-based AOPs and the Fenton process are often considered in landfill-stabilized leachate treatment due to their higher efficiency compared to other AOPs (75). Oxidant $S_2O_8^{-2}$ alone in landfill leachate treatment can reduce 39% COD and 22% NH_3-N, which are difficult to remove by other AOPs. When $S_2O_8^{-2}$ was combined with the ozonation process, the reduction efficiency improved by up to 33% for COD and NH_3-N, and a higher reduction efficiency was obtained compared to fentozone without residual production (75,76).

It seems that according to the approach of producing products in society and the nature of the production leachates, the use of advanced chemical oxidation processes, especially in the removal of hard degradable

compounds and micro-pollutants, is inevitable. Removing leachate color and comparing the effect of each of the investigated processes in this field in the reviewed studies shows that: Fenton > Photo-Fenton > O_3/H_2O_2 > ozonation (O_3) > O_3/UV/H_2O_2 > O_3/UV > O_3/catalyst > UV/TiO_2 > PS (Figure 3).

The ozone-based process was 40 to 95% effective in COD removal with COD concentrations ranging from 2000 to 26000 mg L^{-1} and the Fenton process was 70 to 90% effective in COD removal with COD concentrations ranging from 1855 to 8894 mg L^{-1}. The treatment efficiency of the Fenton process is better than the ozone-based process (34,77-79). The Fenton process is technologically simple, there are no mass transfer limitations (homogeneous adults), and both iron and H_2O_2 are relatively inexpensive and non-toxic. Due to the need for the Fenton process for low pH, it is necessary to modify this parameter (80).

Factors such as catalyst type, oxidant dosage, pH, and operational parameters will be taken into account to determine the cost-effectiveness of different AOPs (81). By comparing AOPs based on these indicators, the present study aimed to provide insights into the most efficient and economically viable options for leachate treatment (65). The findings will help decision-makers and practitioners select the most suitable AOPs for their specific leachate treatment requirements (82). Different technologies may use different catalysts or combinations of catalysts. For example, the Fenton and Photo-Fenton processes utilize iron-based catalysts, while UV/TiO_2 relies on titanium dioxide as the catalyst. The choice of catalyst can affect the efficiency and effectiveness of the treatment process, as well as the overall cost (83).

The pH of the leachate plays a crucial role in determining the performance of the treatment technologies (84). Some processes, like Fenton and Photo-Fenton, are highly pH-dependent and require specific pH ranges for optimal performance (85). Other technologies, such as UV/TiO_2, may have a wider pH operating range (86). The Fenton process requires low pH conditions for optimal performance. Therefore, it is necessary to modify the pH parameter to achieve the desired treatment efficiency (87).

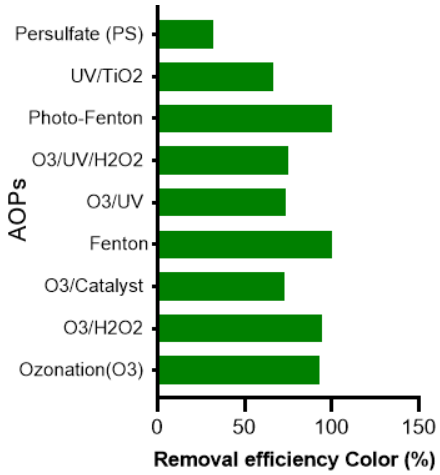

Figure 3. Comparison of AOP_s in terms of color removal efficiency

The amount of oxidant required for effective treatment varies among different technologies. Ozone-based processes, such as O_3 and O_3/catalyst, typically require higher oxidant doses compared to other technologies (88,89). The oxidant dose can affect the cost of the treatment process, as well as potential by-product formation and safety considerations (90). To compare these technologies, it is essential to collect site-specific data, including leachate characteristics, treatment objectives, and desired treatment capacity. Additionally, conducting a comprehensive evaluation of the operational and economic factors associated with each technology can help determine their feasibility and potential cost-effectiveness in leachate treatment (91). This analysis should consider factors such as capital costs, operating costs, energy consumption, maintenance requirements, and potential by-product formation (92).

Discussion

AOPs mechanism in leachate treatment

For an easy understanding of how AOPs work in leachate treatment, an overview of the mechanism involved in the production of oxidants in both direct and indirect forms and the oxidation and degradation of leachate organic matter is shown in Figure 4. The performance of AOPs is increased by optimally manipulating the variables related to AOPs - the type of oxidant and its concentration (75). The organic compound is the treatment target in the AOP process, the basic reaction in AOPs is depicted in Eq. (1).

$$Oxidant + organic\ compound \rightarrow oxidation\ byproducts \rightarrow CO_2 + H_2O \quad (1)$$

Oxidants react with organic compounds in the environment and break them down into intermediate products and turn them into water and carbon dioxide (Figure 4). The most important and common oxidants used in AOPs are ozone (O_3), hydrogen peroxide (H_2O_2), and PS ($S_2O_8^{2-}$). Figure 1 shows the oxidants and radical species involved in AOPs and their oxidation potential. Although different mechanisms are involved in AOPs, in general, the mechanisms of AOPs depend on the oxidant and initiator involved. If the oxidants and initiators are the same in different oxidation processes, similar mechanisms might be involved (75). In the rest of this article, various mechanisms of advanced chemical oxidation processes are discussed independently in each section with a brief overview.

Ozonation (O_3)

Ozone oxidation depends on the composition and pH of leachate and can be done by molecular O_3 reactions or $^{\bullet}OH$ radical reactions (12). At high pH (pH 8 to 9), the

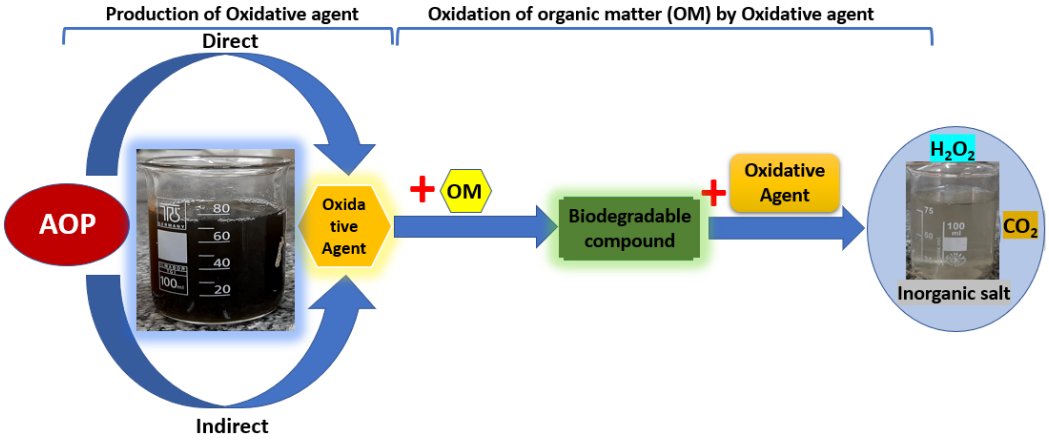

Figure 4. General mechanism of AOPs in leachate treatment

ozonation performance can be increased because this pH favors the reaction between the hydroxyl ion (•OH) in aqueous solution and O_3, which leads to the formation of hydroperoxide ion ($\bullet HO_2^-$), which is an initiator to the production of •OH (75,93,94). A set of the following reactions shows the mechanism of ozonation in leachate treatment:

$$\bullet HO_{2^-} + O_3 \rightarrow \bullet OH + O_{2^-} + O_2 \quad (2)$$

$$\bullet HO_{2^-} + O_3 \rightarrow \bullet OH + O_{2^-} + O_2 \quad (3)$$

$$O_3 + \bullet OH \rightarrow \bullet O_{2^-} + \bullet HO_2 \quad (4)$$

$$O_3 + \bullet O_{2^-} \rightarrow \bullet O_{3^-} + O_2 \quad (5)$$

$$\bullet HO_3 \rightarrow \bullet oH + O_2 \quad (6)$$

$$\bullet HO_3 \rightarrow \bullet oH + O_2 \quad (7)$$

$$\bullet oH + O_3 \rightarrow \bullet OH_2 + O_2 \rightarrow \bullet HO_4 \quad (8)$$

$$\bullet HO_2 \leftrightarrow \bullet O_2 + H^+ \quad pka = 4.8 \quad (9)$$

$$\bullet HO_2 \leftrightarrow \bullet O_2 + H^+ \quad pka = 4.8 \quad (10)$$

$$\bullet HO_4 \leftrightarrow \bullet HO_4 \rightarrow H_2O_2 + 2O_3 \quad (11)$$

$$\bullet HO_4 \leftrightarrow \bullet HO_3 \rightarrow H_2O_{2^-} + O_3\ O_2 \quad (12)$$

Catalytic ozonation can oxidize unsaturated conjugated bonds and aromatic structures in organic materials by producing active radicals, such as hydroxyl radical (•OH) (95). During catalytic ozonation, the hydroxyl radical produced by O_3 combines with oxygen, and H_2O_2 radical is produced during the reaction, while the unpaired electrons are trapped by surface-adsorbed oxygen on the catalysts to produce superoxide radicals. In addition, active metal components on the surface of the catalyst can become synergistic, which can maintain the activity of the catalyst and remove pollutants (96). In the ozonation process, the COD reduction mainly depends on the presence of molecular O_3, and the presence of hydroxyl radicals is essential. The amount of COD reduction and leachate purification in the ozonation process depends on its time and pH. In alkaline solutions (up to about 11), due to the difference in the amount of O_3 transferred to the leachate and the subsequent increase in the rate of O_3 decomposition and the increase in hydroxyl radicals, COD decreases. In alkaline pH, hydroxyl radicals are non-selective towards large unsaturated and complex molecules. In leachate with less alkalinity (about pH 8), a strong O_3 oxidant reacts selectively with leachate (97). The ozonation process (O_3) is a promising and suitable technology for removing organic materials resistant to biological degradation with high efficiency and without secondary pollution (30). Organic components of solid waste leachate can be decomposed and even mineralized by O_3 and/or highly oxidative hydroxyl radicals (•OH) from O_3 decomposition (98). The O_3 process is enhanced by a dose of H_2O_2 (Peroxone process) and increases the formation of •OH. Advanced chemical oxidation using O_3 is still expensive as a final treatment process due to the high energy consumption for O_3 production (99). Therefore, effective O_3 dosage is very important to reduce operating costs (100). The ozonation process consists of two primary reactions, corresponding to the instantaneous phase, in which applied O_3 dose (AOD) < initial O_3 demand (IOD), and the gradual phase, in which AOD > IOD, respectively. Due to the different reaction mechanisms, the rate of mass transfer and O_3 decomposition in the two stages is different. Therefore, the O_3 efficiency and •OH yield in different ozone-based AOPs may vary significantly during ozonation (100). In the combined treatment using ozonation as a pre-treatment, and then, adsorption on zeolite in improving leachate purification in COD, NH_4-N, and color removal increased to 82%, 75%, and 92% after using adsorption as a post-treatment. The results using ozonation or surface adsorption process alone showed good removal for the target parameters removal of 89% for color and 45% for COD was achieved only with ozonation (101).

O_3/H_2O_2 (Peroxone)

In the Peroxone process, using ozonation with the addition

of a strong oxidant, H_2O_2 increases the conversion of O_3 to •OH through the O_3/H_2O_2 reaction (102,103). In the Peroxone process, apart from O_3 mechanisms, additional reactions are involved as follows:

$$H_2O_2 + H_2O \rightarrow HO_{2^-} + H_3O^+ \quad (13)$$

$$H_2O_2 + H_2O \rightarrow HO_{2^-} + H_3O^+ \quad (14)$$

$$\bullet OH + \bullet HO_2 \rightarrow H_2O + O_2 \quad (15)$$

The study of Wang et al comparing the reduction of macro- and micropollutants from biologically treated landfill leachate with single ozonation, O_3/H_2O_2, and catalytic ozonation showed that the role of catalytic ozonation is mainly to increase the direct oxidation of O_3, while the O_3/H_2O_2 process is to promote indirect oxidation (104). The highest synergistic effect was obtained by combining O_3 with hydrogen to treat waste landfill leachate (29). The O_3/H_2O_2 process considerably enhanced the leachate biodegradability from 17% to 79% after a 3-hour reaction period (105). The production of hydroxyl radicals (•OH) by combining O_3 with H_2O_2 is favorable and a synergistic effect can be expected for the removal of organic matter (106). The ozone-based treatment, O_3/H_2O_2, did not meet the effluent that can guarantee the legal COD discharge limit (< 150 mg L^{-1}) after further biological treatment (29).

The research conducted in the field of operational parameters in AOPs revealed that higher doses of O_3 and a higher pH generally increase the reduction of efficiency in the AOP of O_3/H_2O_2. The parameters investigated included O_3 dose (0.5–3 mg L^{-1})), H_2O_2 dose (O_3:H_2O_2 = 1:3-3:1 mass ratio), and pH (6.5-8.5). It was found that the addition of H_2O_2 plays an important role in accelerating the conversion of O_3 to OH radicals, thereby, facilitating faster removal of ozone-resistant micropollutants. However, it was observed that the reduction of micropollutants decreased with higher oxidant concentration during conventional ozonation due to competitive O3-consuming reactions (107).

O_3/catalyst process

Although ozonation and AOPs are parts of the chemical and physical method that has recently received attention, and ozonation of leachate reduces high-strength organic molecules to smaller molecules and improves biodegradability, however, the use of O_3 alone is ineffective and needs to be combined with a catalyst to increase biodegradability and break down complex organic structures in samples (108,109). Landfill leachate treatment can be improved by adding catalytic materials in a column reactor with O_3 or peroxone systems and by increasing the contact surface of the landfill leachate. In the treatment of landfill leachate by ozonation using a catalyst, the capacity to remove organic compounds increases directly and indirectly by increasing the contact surface of the leachate with catalytic materials. Ozone is used to treat landfill leachate to remove organic compounds. Ozone alone, peroxide, O_3/UV, O_3/Fenton, O_3/GAC, catalytic O_3 with Fe^{2+}, Al^{3+}, $S_2O_8^{2-}$, and TiO_2 have been used (110). The results of investigating the catalytic effect of Co^{2+}, Ni^{2+}, Cu^{2+}, Mn^{2+}, Zn^{2+}, Cr^{3+}, and Fe^{2+}ions in the O_3/catalyst process showed that $CO^{2+}$and Mn^{2+}have the most catalytic activity for the decomposition of oxalic acid in solution, while the catalytic effect of the studied ions on the speed of the decomposition of formic acid is negligible (111). The testing of three types of activated carbon (AC), expanded perlite (EP), and titanium dioxide (TiO2) catalysts combined with O3 at a gas concentration of 80 g/m3 for treating the leachate produced at the Jabal Chakar landfill site near Tunis, the capital of Tunisia, was conducted. This work has shown a reduction (about 45%) in COD and an increase in biodegradability (BOD_5/COD) from 0.1 to 0.34. A catalyst concentration of 0.7 g L^{-1} was optimal for leachate treatment (112). The combined performance of O3 and ZrCl4 has been shown to effectively remove color (70-75%), ammonia (53-69%), and COD (33-48%). The biodegradability results also improved from 0.07 to 0.08 for Alor Pongsu Landfill Site and 0.05 to 0.11 for the Pulau Burung Landfill Site (101).

Fenton process (H_2O_2/Fe_2^+)

The Fenton process is a reaction between hydrogen peroxide (H_2O_2) and iron ion (Fe^{2+}), which produces hydroxyl radical (•OH) (113,114). In the Fenton process, metals such as iron are responsible for the formation of hydroxyl radicals through the Fenton reaction. Fe (II) ions readily get oxidized into Fe (III) in the presence of excess H_2O_2. The mechanism of this reaction is shown in Eqs. 16 to 22 (70,115).

$$H_2O_2 + Fe^{2+} \rightarrow Fe^{3+} + \bullet OH + OH \quad (16)$$

$$H_2O_2 + \bullet OH \rightarrow H_2O_2 + \bullet HO_2 \quad (17)$$

$$H_2O_2 + \bullet OH \rightarrow H_2O_2 + \bullet HO_2 \quad (18)$$

$$Fe^{2+} + \bullet OH \rightarrow Fe^{3+} + OH^- \quad (19)$$

$$Fe^{2+} + \bullet HO_{2+}H^+ \rightarrow Fe^{3+} + H_2O_2 \quad (20)$$

$$Fe^{2+} + \bullet HO_{2+}H^+ \rightarrow Fe^{3+} + H_2O_2 \quad (21)$$

$$\bullet HO_2 \rightarrow H_2O_2 + O_2 \quad (22)$$

Hydrogen peroxide plays an important role in treating Fenton's process. The concentration of H_2O_2 depends on the stoichiometric ratio of H_2O_2 and the initial COD of the leachate (97). The results show that refractory aromatics and carboxylic structure in leachate are decomposed

into aliphatic compounds after Fenton oxidation. Fulvic-like and humic-like species, both of which can be removed during Fenton treatment. Although refractory macromolecular materials are broken by Fenton oxidation. However, studies show that these substances cannot be completely removed by Fenton oxidation (35). More Fe (II) catalysts can speed up the chemical oxidation process and increase the removal of organic matter through sedimentation. But using too much iron (II) produces too much sludge, and thus, increases costs (35,116,117). Fenton's reagent with the combination of H_2O_2 and iron salt can eliminate organic pollutants in all types of wastewaters. The Fenton process is carried out at an ambient temperature, it is easy to work with, and there is no problem in terms of safety (117-119). The use of the Fenton process reduces the BOD_5 and COD of landfill leachates, and many studies have reported the high efficiency of the Fenton process in removing leachate organic matter (47,120).

The study on the effect of pH on the Fenton process demonstrated a significant impact of this parameter on the reaction. It was found that higher pH levels led to a decrease in COD removal efficiency, with a reduction of 15.8%. This decrease can be attributed to the precipitation of iron at higher pH values, which hinders its interaction with H_2O_2, and subsequently, reduces the production of the •OH radical (121). A study analyzing peer-reviewed publications found that the optimal pH range for Fenton oxidation of raw, biological, and coagulation-treated leachates is 2.5–4.5, with a median value of 3.0. For biologically treated leachates, the optimal range is 2.5–6.0, with a median value of 4.2. This highlights the importance of pH control in Fenton oxidation processes (122).

Photolytic ozonation (O_3/UV)

In photolytic ozonation (O_3/UV), O_3 is activated by absorbing ultraviolet light at a wavelength of 254 nm and undergoes photolysis to produce H_2O_2, and then, •OH (123). The mechanism of the O_3/UV system is shown in Eqs. 23 to 25 is the same as the ozonation mechanism with additional steps.

$$O_3 + H_2O \xrightarrow{h\nu} O_2 + H_2O_2 \quad (23)$$

$$H_2O_2 \xrightarrow{h\nu} 2\bullet OH \quad (24)$$

$$2O_3 + H_2O_2 \rightarrow 2\bullet OH + 3O_2 \quad (25)$$

The O_3/UV_C oxidation process among ozone-based AOPs processes (O_3/H_2O_2, O_3/UV_C, O_3/H_2O_2/UV_C) for treating mature municipal landfill leachate with 86% and 43% for DOC and COD removal and 91%, respectively, increased biodegradability was the best one, resulting in the highest synergistic effect (105). Photodegradation of O_3 by UV_C light has been found to increase HO• production and can also cause H_2O_2 formation (124). The produced H_2O_2 can further contribute to the production of •OH by UV_C photolysis. Therefore, indirect oxidation reactions can be dominant for the O_3/UV_C process (29).

Photolytic peroxide (O_3/H_2O_2/UV) and (H_2O_2/UV)

During the UV/H_2O_2 process, H_2O_2 is activated by ultraviolet (UV) radiation and produces more reactive OH radical species, which further oxidize organic matter in the waste leachate (125,126). Hydroxyl radicals are formed by hemolytic splitting of the oxygen–oxygen bonds of H_2O_2 by UV light at a wavelength (λ) of 200 to 300 nm (Eqs. 26 to 29) (12,127,128).

$$H_2O_2 + \bullet OH \rightarrow \bullet HO_2 + H_2O \quad (26)$$

$$H_2O_2 + \bullet OH \rightarrow \bullet HO_2 + H_2O \quad (27)$$

$$\bullet OH + \bullet OH \rightarrow H_2O_2 \quad (28)$$

$$\bullet OH + \bullet OH \rightarrow H_2O_2 \quad (29)$$

In the use of AOPs based on hydroxyl radicals (UV/H_2O_2) to remove persistent organic pollutants (POPs) from wastewater, hydroxyl radicals (•OH) react with many organic chemicals at near diffusion-controlled rates. Applying O_3/H_2O_2 is too expensive for many landfills that treat leachate of various ages (129). pH plays an important role in removing color in the H_2O_2/UV process, and this process is more effective in terms of color change in an acidic environment (125). The biodegradability of leachate was increased by 79% and 85% using each of the O_3/H_2O_2 and O_3/H_2O_2/UVC processes, respectively (29). The UV/H_2O_2/O_3 process is an efficient method against micropollutants (130). UV/H_2O_2 and UV/H_2O_2/O_3 technologies are efficient in the oxidation of organic matter and even complete mineralization of some pollutants and reduction of chemical oxygen demand (COD), toxicity, and TOC (131).

Photo-Fenton and photo-Fenton like

Photolytic Fenton or photo-Fenton is an improved version of the Fenton system by utilizing UV irradiation. Of photolyzed UV (UV > 300 nm) to photolyzed ion (Fe^{3+}) complexes, it is used to regenerate Fe^{2+}, and thus, reinitiate the Fenton reactions. The mechanism of photo-Fenton is shown in Eq. (30) (75,127,132). Fenton-like processes are modified techniques using heterogeneous catalysts except for iron salts and H_2O_2 (external addition or in situ production) (133-135), which have the same mechanism as the photo-Fenton process.

$$H_2O_2 + Fe^{2+} \xrightarrow{h\nu} Fe^{3+} + \bullet OH + OH^- \quad (30)$$

In the photo-Fenton process (UV/Fe^{2+}/H_2O_2), iron salt and H_2O_2 are the two main chemicals that determine the

costs and efficiency of the operation, and H_2O_2, Fe^{2+}, Cu^{2+}, and UV light, and temperature are related operational variables (33). In the photo-Fenton process, due to the combination of ultraviolet rays with the traditional Fenton reaction accelerates the reduction of dissolved $Fe^{3+}$to Fe^{2+}. During the treatment of landfill leachate by Fenton-like and photo-Fenton processes under optimal conditions, COD removal of 69.6%, 65.9%, and 83.2% can be achieved, respectively (136).

Photocatalysis (UV/TiO_2)

The mechanism TiO_2/UV lies in the production of OH free radicals by inducing electron transformation using UV illumination. Its mechanism is similar to H_2O_2/UV, and O_3/UV, with the difference that TiO_2 has priority over others due to its properties including stability in different conditions, its high potential for radical production, and its easy availability and low price (137). The efficiency of the TiO_2 photocatalytic process in removing some resistant organic and inorganic pollutants in landfill leachate is due to certain factors such as the type and mode of pollutant, photoreactors, operational parameters, and environmental dynamics (72). As shown in Eqs. (31 to 34) by absorbing UV light (< 380 nm) in TiO_2, electrons are promoted to move from the valence band (VB) to the conduction band (CB) to produce electron-hole pairs (e^-/h^+). Electrons mainly reduce molecular oxygen to superoxide radical anions. These radicals oxidize a wide range of organic pollutants to harmless minerals such as CO_2, H_2O, and other minerals (138-141). In the optimal conditions of the photocatalyst process with 1 g/L TiO_2, COD was removed by 82%. The rate of removal of COD increased to 85% with a dose of 1.5 g/L during the irradiation time of 2 hours (72). The increase in the BOD_5/COD ratio from 0.09 to 0.39 after 72 hours indicates an enhancement in the biodegradation ability of the leachate through photocatalysis (UV/TiO_2) (55).

$$TiO_2 + (hvB) + HO^- \rightarrow + TiO_2 + HO^\circ \quad (31)$$

$$TiO_2 + (hvB) + HO^- \rightarrow + TiO_2 + HO^\circ \quad (32)$$

$$R + HO^\circ \rightarrow R(HO^\circ) \rightarrow R1 + R2 \rightarrow Inorganic\ substances\ (i.e. CO_2 + H_2O) \quad (33)$$

$$R + HO^\circ \rightarrow R(HO^\circ) \rightarrow R1 + R2 \rightarrow Inorganic\ substances\ (i.e. CO_2 + H_2O) \quad (34)$$

Studies have demonstrated that the optimal dosage of TiO_2 catalyst is critical for efficient photocatalytic degradation processes. This is because an optimal dosage can enhance the generation of reactive species, such as hydroxyl radicals, and increase the photocatalytic activity. However, excessive dosage can lead to aggregation and decreased light penetration, resulting in a lower photocatalytic efficiency (72,142,143). A dosage of 1 g/L^{-1} enhances electron/hole pair production and OH radical formation, but reduces light penetration and photodegradation rates. This is crucial for efficient phenol removal in TiO_2/UV processes, especially at an initial phenol concentration of 0.51 mM and pH 6.8. Determining the ideal dosage is essential for optimal TiO_2-based photocatalysis (142).

PS advanced oxidation process

In the photolytic PS system, SO_4^- is produced through the photolysis of $S_2O_8^{2-}$ by the irradiation of ultraviolet rays as the initiator of the reaction. Also, in the continuation of the reactions, the decomposition of $S_2O_8^{2-}$ is done through R (•OH) resulting from the photolysis of H_2O_2. Hydrogen peroxide is the result of the reaction of $S_2O_8^{2-}$ with water molecules (75,144). The mechanisms of this system include H_2O_2/UV reactions and initiator reactions as follows:

$$S_2O_8^{2-} + 2H_2O \rightarrow 2\,SO_4^- + H_2O_2 \quad (35)$$

$$S_2O_8^{2-} + 2H_2O \rightarrow 2\,SO_4^- + H_2O_2 \quad (36)$$

$$H_üO \xrightarrow{hv} 2{\bullet}OH \quad (37)$$

PS AOP is an advanced technology in the field of landfill leachate treatment. SR-AOP appears to be more advantageous over hydroxyl radical (•OH)-based advanced oxidation processes (HR-AOPs) because •OH almost does not oxidize ammonia (9,62). In the UV solar/O_3/H_2O_2/$S_2O_8^{-2}$ process, the interaction of $S_2O_8^{-2}$ and •OH leads to the oxidation of more organic compounds (63). Heat-activated PS process may be an alternative technology for COD and inert COD removal from leachate nanofiltration concentrate (64). The biodegradability (BOD_5/COD) ratio improved from 0.034 to 0.29 following O_3/ $S_2O_8^{2-}$ (144). Under the conditions, including the initial humic acid concentration of 200 $mg\ L^{-1}$, PS dose of 25 mmol/L, and initial pH of 4, the maximum UV_{254} and TOC removal rates were 89.62% and 76.17%, respectively. Therefore, the UV/PS system is effective in removing humic acid from landfill leachate and has high efficiency (145).

Optimizing leachate treatment with AOPs

AOPs are effective for treating complex organic pollutants in leachate treatment (146,147). They generate highly reactive hydroxyl radicals (•OH) for efficient degradation of organic contaminants. Understanding the composition and concentration of pollutants in leachate is crucial for selecting the best leachate treatment process using AOPs (65,148). Factors such as reaction kinetics, operational parameters, energy consumption, and environmental impact should be considered. AOPs can degrade POPs, refractory compounds, and specific contaminants (149,150). Operational parameters like pH, temperature, catalyst type, and oxidant dosage also play a role. Energy consumption and cost are essential for practical implementation (151). Environmental impact should be assessed through a comprehensive life cycle assessment. Combining laboratory studies, pilot-scale trials, and

modeling can provide valuable insights for selecting the best treatment process (152,153).

In this study, a comprehensive analysis was conducted to evaluate the performance and technical and economic capabilities of different AOPs in leachate treatment. The analysis compared AOPs based on indicators such as energy consumption, cost-effectiveness, and technical feasibility. The results of the evaluation can be seen in Table S2.

The energy consumption of AOPs will be assessed by considering factors such as oxidant generation, reaction conditions, and overall process efficiency (81,154). This analysis will help identify AOPs that are more energy-efficient and have lower energy requirements for leachate treatment (154). Cost-effectiveness will be evaluated by considering the capital and operational costs associated with implementing and maintaining AOPs (155,156).

To conduct a precise economic and operational analysis of the AOP in leachate treatment, it is crucial to gather site-specific data and perform calculations based on the unique conditions and requirements of the leachate treatment facility (157,158). This entails obtaining information on factors such as leachate volume, composition, treatment goals, and the desired capacity of the AOP (137). Using these data, accurate cost estimates can be generated, allowing for a comprehensive evaluation of the economic feasibility and potential cost savings associated with implementing AOPs in leachate treatment (159-162).

Conclusion

AOPs are effective methods for treating landfill leachate by breaking down stubborn organic matter using hydroxyl radicals. These processes can be categorized as non-photochemical or photochemical, both utilizing free radicals as oxidizing agents. Among the various AOP techniques, UV-based methods are particularly recognized for their ability to remove micropollutants and pathogens by generating hydroxyl radicals.

Different AOPs exhibit varying levels of effectiveness in leachate treatment. In terms of COD removal, the following processes demonstrated the highest efficiencies: O_3/UV/H_2O_2, Photo-Fenton, UV/TiO_2, Fenton, PS, O_3/UV, O_3/H_2O_2, O_3/catalyst, and ozonation (O_3), with a maximum efficiency of 83.75%. The removal of leachate color through chemical oxidation processes ranged from 32% to 100%, depending on the leachate composition and process employed. Hybrid AOP methods appear to be more promising and effective compared to individual AOPs.

Leachate characteristics, technical feasibility, discharge standards, cost-effectiveness, regulatory compliance, and environmental impacts should be considered when selecting the most suitable AOP for leachate treatment. AOPs, especially hybrid approaches, play a crucial role in degrading persistent organic compounds in waste leachate. By combining different AOP methods, hydroxyl radicals can be efficiently generated, treatment parameters optimized, and persistent organic compounds effectively removed. However, the efficiency of AOPs can vary depending on treatment parameters and leachate composition. Further research is necessary to enhance the treatment process and improve AOP efficiency in waste leachate treatment.

In conclusion, AOPs, particularly the hybrid approach, have significant potential for effectively treating waste leachate and removing persistent organic compounds. However, further optimization and research are required to enhance their efficiency in specific treatment scenarios.

Strengths, limitations, and future research

In this article, the authors conducted a review of common and effective AOPs used in leachate treatment. They emphasize the need for further research on various types of photocatalysis processes specifically designed for treating leachate under sunlight, focusing on the charge transfer mechanism. Although electrochemical oxidation was not investigated in this study due to its high energy consumption and the formation of chlorinated organic substances, it is acknowledged as a promising and powerful technology for landfill leachate treatment. Additionally, the authors suggest exploring other oxidants such as potassium permanganate ($KMnO_4$), peroxymonosulfate (PMS), chlorine (Cl), and novel chemical oxidation methods for leachate treatment, warranting further investigation and study.

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Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethical issues

This study was approved by the Ethics Committee of the Mazandaran University of Medical Sciences (Ethical code: IR.MAZUMS.REC.1401.377). The authors certify that all data collected during the study are as stated in the manuscript, and no data from the study has been or will be published separately elsewhere.

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Supplementary files

Supplementary file 1 contains Figures S1-S2 and Tables S1-S2.

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