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Landfill leachate treatment using a combined method of coagulation, flocculation, advanced oxidation, and extended aeration

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Abstract

Background: Modifying and enhancing treatment methods is essential to meet effluent standards for treating landfill leachate. This study investigated the treatment of municipal solid waste leachate (MSWL) using coagulation, flocculation, advanced oxidation, and extended aeration processes.

Methods: The effects of different coagulant doses and pH values on coagulation processes were compared. The treatment procedure was analyzed to determine the impact of varying concentrations of potassium persulfate ($K_2S_2O_8$) and hydrogen peroxide (H_2O_2) on the results after coagulation with $FeCl_3$. The extended aeration process's biological stages were studied using a sludge retention time (SRT) of 23 days and the effects of hydraulic retention time (HRT) of 18 and 36 hours.

Results: The experimental results show that in the pH range of 5–8, the lower the pH value, the higher the treatment efficiency. The addition of 0.8 g L^{1-} $FeCl_3$ can achieve a 57% removal of chemical oxygen demand (COD). The addition of 2.5 g L^{1-} $K_2S_2O_8$ and 1.5 g L^{1-} H_2O_2 with UV-C (15 W) for 70 minutes at pH 7 can effectively remove 86% of COD. Activated sludge extended aeration can attain an 88% removal of COD under optimal operating conditions (HRT = 36 hours, SRT = 23 days, and aeration = 36 hours). The studied hybrid process with the efficiency of 99%, 98%, 95%, 87%, and 83% removal of COD, biochemical oxygen demand (BOD), total suspended solids (TSS), turbidity, and total Kjeldahl nitrogen (TKN), respectively, is suitable for leachate treatment.

Conclusion: This study showed that flocculation-coagulation followed by the advanced oxidation process (AOP) and extended aeration can be an efficient and promising treatment method for MSWL.

Keywords: Potassium persulfate, Solid waste, Hydrogen peroxide, Flocculation, Leachate

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Introduction

Leachate is a very thick, resistant, and toxic liquid that is produced from the physical, chemical, and biological changes of solid waste in landfills, incinerators, compost plants, and transfer stations (1-4). Leachate is produced from waste as a result of water percolating through the waste materials, extracting various contaminants and pollutants as it passes through (5,6). The sources and origin of leachate can include landfills, composting facilities, waste storage areas, and industrial sites (7). In landfills, rainwater and other liquids come into contact with decomposing waste, extracting dissolved and suspended materials, organic compounds, heavy metals, and other pollutants (2,8). Similarly, in composting facilities, water used in the composting process can leach out soluble organic compounds and nutrients, forming leachate (9,10). Leachate can also be produced in areas where waste is stored, such as waste piles or storage containers, as a result of rainfall or other forms of water infiltration (11). Industrial activities involving the storage or disposal of waste materials can also generate leachate containing various contaminants and pollutants, including chemicals, heavy metals, and organic compounds (5,12). It is important to note that the composition and characteristics of leachate can vary widely depending on the type of waste, the age of the waste, and the environmental conditions (13). Proper management and treatment of leachate are essential to prevent environmental contamination and protect water resources (14). Due to the serious pollution caused by leachate to water resources and the environment, to treat leachate and reduce the risk of pollution caused by it, it is necessary to develop sustainable and efficient leachate treatment techniques to protect the environment and public health (15-17). Surveys show that the per capita production of waste per person in Iran is 0.7 to 1 kg per day (average of 0.75 kg/d), in which organic waste occupies a significant amount (18). The per capita waste production in Mazandaran province, located in the north of Iran, is estimated to be about 1 kg (19-21). In most coastal cities of Mazandaran, due to the disposal of municipal solid waste (MSW) in landfills that do not have a leachate collection and treatment system, leachate easily enters water sources near the disposal site (22,23). Considering the composition of leachate and the hazardous chemicals in leachate, choosing an efficient method for leachate treatment is still an enormous challenge. Recently, the combination of biological processes and physicochemical methods has become very efficient (24). Flocculation-coagulation is widely applied as a pretreatment of waste leachate due to its simplicity and cost-effectiveness (25,26). The strict and ever-increasing environmental requirements require us to use new and more advanced methods to fill the existing gaps to reach today's environmental standards, and one of these methods is advanced chemical oxidation (27). In recent years, they have preferred the application of persulfate-based advanced oxidation methods due to its high reactivity and the creation of sulfate radicals. The sulfate radical provides an easy implementation of reactions with various catalysts (28,29). Sulfate-based advanced oxidation processes (AOPs) are more effective in the degradation of resistant organic and inorganic materials, such as compounds with unsaturated aromatic bonds (30). Persulfate (PS) ions are commonly present in the form of stable sodium ($Na_2S_2O_8$) and potassium ($K_2S_2O_8$) salts, with a high redox potential. These salts are widely utilized in the remediation of water and soil (31). To produce sulfate-free radical ($SO4^{\bullet-}$), persulfate is activated using chemical or thermal methods. According to the sulfate radical oxidation potential (E0 = 2.6 V), it has a high oxidation capacity (32). In the design of AOPs, one of the most important criteria is the stable production of free radicals ($OH^{\bullet}$). Hydroxyl radicals attack these compounds through the abstraction of hydrogen from organic compounds or through the addition of OH radicals to organic molecules (33). In contrast, $SO4^{\bullet-}$ usually converts organic molecules into organic radicals through electron transfer (34).

The selection criteria for leachate treatment processes in a project typically include the effectiveness in removing specific pollutants, the associated costs, operational efficiency, and the available facilities and equipment at the leachate treatment site (35). The cost of the treatment is an important parameter to consider, and different treatment methods are ranked based on their performance in removing specific pollutants and their cost-effectiveness (35,36). The decision-making process for leachate treatment involves balancing technical, economic, and environmental aspects of sustainability (36). Various treatment techniques, including biological, physical/chemical, and hybrid methods are employed to treat landfill leachate, and pre-treatment is often required for heavily contaminated wastewater (37).

In leachate treatment, the coagulation and flocculation stages are crucial for removing suspended particles and soluble pollutants (38,39). The coagulation stage involves adding coagulating chemicals to the wastewater to form flocs that separate suspended solids from the solution. The flocculation stage agglomerates the formed flocs, causing the accumulation of solid particles. These processes are essential for leachate treatment due to their high efficiency in removing suspended particles and soluble pollutants (39,40).

AOPs are used to treat wastewater containing refractory, toxic, or non-biodegradable materials (41-43). AOPs generate hydroxyl radicals ($OH^{\bullet}$) or sulfate radicals ($SO_4^{\bullet-}$) in sufficient quantities to remove traceable organic contaminants and certain inorganic pollutants, or to increase wastewater biodegradability as a pre-treatment before an ensuing biological process (44). Ultraviolet (UV) radiation can activate persulfate and hydrogen peroxide (H_2O_2), leading to the formation of sulfate and hydroxyl radicals (17,45). These radicals are powerful oxidants that can rapidly decompose organic compounds into smaller and less harmful molecules. This process is particularly effective in breaking down complex organic molecules that are resistant to conventional treatment methods (46). The synergistic effect of UV-PS/H_2O_2 in the treatment of wastewater leads to the degradation of organic pollutants, reduction of color, and elimination of odor. It also aids in the removal of pathogens and pharmaceutical residues, making it an efficient and versatile treatment option for wastewater (17). In general, the UV-PS/H_2O_2 process has shown great promise in leachate treatment by effectively reducing the concentration of various pollutants and improving the overall quality of the effluent (17,47).

The extended aeration-activated sludge (EAAS) process is a common method for leachate, wastewater, and effluent treatment (48). This process utilizes activated sludge, air, and microorganisms to remove organic matter and nitrogen from wastewater (49,50). In the EAAS process, wastewater is introduced into large tanks containing activated sludge microorganisms, to which air is supplied (51). This process relies on providing air and nutrients to the microorganisms to create an optimal environment for their activity. The microorganisms decompose the organic matter present in the wastewater using oxygen from the air, converting pollutants into non-organic substances (48,50,51). This process is effective due to the prolonged contact time between wastewater and microorganisms (long-duration aeration), providing the possibility of complete purification. Additionally, this process produces activated sludge, which can be used as a food source for microorganisms in subsequent treatment processes. As a result, the EAAS process is highly regarded for its efficiency in pollutant removal, high-quality activated sludge production, and facilitation of complete wastewater treatment. It is a notable treatment process for both wastewater and leachate due to its effectiveness (17,48,52).

In the treatment of landfill leachate, AOPs have been used in combination with other methods, such as coagulation and membrane bioreactor integration, to achieve high removal efficiencies of pollutants like aluminum, chemical oxygen demand (COD), suspended solids, and total organic carbon. Hybrid physical/chemical methods, including AOPs, have also been proposed to improve removal efficiency and decrease energy consumption (17,37,53).

Improving and optimizing treatment approaches is crucial for meeting effluent standards in the treatment of landfill leachate. Therefore, the primary aim of this study was to analyze the physicochemical properties of urban waste leachate and to design a laboratory-scale Batch-flow leachate treatment system (BFLTS) based on sulfate-hydroxyl radicals for municipal solid waste leachate (MSWL) treatment. This research investigated the application of coagulation, flocculation, advanced oxidation, and extended aeration processes in the treatment of MSWL.

Materials and Methods

Chemicals and samples

Aluminum sulfate ($Al_2(SO_4)_3$), Ferrous sulfate heptahydrate ($FeSO_4·7H_2O$), ferric chloride ($FeCl_3$), concentrated sulfuric acid (H_2SO_4), sodium hydroxide (NaOH), and Potassium hydrogen phthalate (KHP) were obtained from Merck (Germany). Potassium persulfate ($K_2S_2O_8$), H_2O_2, ammonium chloride (NH_4Cl), and sodium thiosulfate ($Na_2S_2O_3$), were purchased from Sigma–Aldrich (USA). Reagents of technical grade and analytical grade were used. Preliminary tests showed that the prepared leachate decays rapidly. For precise control of the loading level, the effectiveness of the treatment system, and the progressive changes in the organic loads, synthetic leachate (SL) (a mixture of trace metals and nutrients in distilled water) was prepared for daily laboratory utilization.

Actual leachate was collected from a municipal waste landfill of Qaem-Shahr city in northern Iran during the dry (summer) and wet (winter) seasons. The pH of the leachate samples was immediately determined at the sampling site with a portable pH meter. Samples from each of the leachate accumulation sites in these locations were collected and filled in high-density polyethylene containers (54), which were previously washed, dried, and prepared, and their lids were covered with clean aluminum foils, from the beginning of the experiments in the laboratory, it was kept in the dark and in a refrigerator at a temperature of 4 degrees Celsius. Before testing, the samples were removed from the refrigerator and placed at room temperature for about 2 hours for conditioning (55).

Analytical methods

The physicochemical characteristics of leachate were determined based on the standard methods for testing water and wastewater (56). The COD analysis was done using the closed reflux method and colorimetric method No. 5220D. DR 2800-HACH spectrophotometer was used for colorimetry and reading the absorption of the contents of the vials at a wavelength of 600 nm. Biochemical oxygen demand (BOD) concentration was determined using the method of Method No. 5210B. Total Kjeldahl nitrogen (TKN) was measured using Method 4500-Norg C. Total suspended solids (TSS) were determined according to Method No. 2540B. The pH parameters were determined using a HI2211pH/ORP meter, turbidity in a HACH-2100P, and conductivity with AQUALYTIC-Sens Direct Con200. Coagulation studies were performed, on the leachate with jar test equipment (Jar Tester Phipps & Bird Stirrer Model-7790-402). Leachate was analyzed for metal content using inductively coupled plasma mass spectrometry (ICP-MS, 7500 Agilent).

Design of the BFLTS

Leachate treatment studies in the stages of coagulation-flocculation, sand filtration, AOPs, and extended aeration were performed. Once the optimal experimental conditions were determined for each process, all subsequent steps were conducted using landfill leachate under these optimal conditions. The experiments were performed in batch mode at a laboratory scale.

Leachate pretreatment was done, with a coagulation-flocculation process performed in the Jar Test Apparatus equipped with 6 beakers of 2.5 L each. The sand filtration was made from a polyvinyl chloride (PVC) column (height: 60 cm; Ø 5.2 cm) equipped with an outlet valve. The column was filled with 700 g of sand from top to bottom: (i) a 30 cm layer of a mixture of filter sands (Ø 0–3 mm); (ii) a 10 cm layer of thin gravel (Ø 5–10 mm); and (iii) a 10 cm layer of coarse gravel (Ø 10–20 mm). The photooxidation processes were carried out in the 2 L laboratory reactor equipped with a UV lamp by Philips. Due to the high temperature caused by the UV lamp, the system was placed inside a 4 L cylinder with a larger diameter filled with water to regulate the temperature using a (water) coolant. The sample temperature was kept constant using a thermometer in the laboratory environment. Air was continuously supplied to the reactor chamber to mix the reactor contents and provide the oxygen necessary for photolysis. A laboratory-scale EAAS treatment system was built. The aeration tank (AT) was rectangular and had an effective volume of 4 L. It was connected to a 1 L cone-shaped settling tank (ST). Air was blown into the leachate through a pump at the AT, and distributed evenly using two diffuser stones. A baffle was constructed at the head of the AT to form a small area for feeding recycled effluent. Figure 1 shows a schematic of the different stages of leachate treatment.

MSWL treatment using BFLTS

An appropriate chemical coagulant was selected based on the characteristics of the raw leachate, COD, and turbidity removal efficiency using coagulation/flocculation tests in a jar test using aluminum sulfate, iron sulfate, and iron chloride as chemical coagulants at concentrations of 0.8, 1.5, and 2 g L^{-1} and pH at different values within the optimum range of each coagulant: $Al_2(SO_4)_3$ from 6 to 9; $FeSO_4.7H_2O$ from 8 to 10, and $FeCl_3$ from 5 to 8 have been achieved. These concentrations were selected based on previous studies (57,58). The pH of the leachate by adding the right amount of concentrated, 5 N H_2SO_4 and/or 5 N NaOH was adjusted. Appropriate contact times including 2 minutes of rapid mixing at 150 rpm for coagulation, 30 minutes of slow mixing at 50 rpm for flocculation, and 60 minutes of settling to promote solids and sedimentation, were applied. The flocculation-coagulation process was carried out under optimal conditions, and the produced effluent was filtered through a sand filter to remove the residual floc before the AOPs.

In this study, the advanced chemical oxidation process with UV-PS/H_2O_2 and Heat-PS/H_2O_2, each separately, was used to determine the effects of independent variables on the removal of desired parameters in the treatment of the leachate. In 500 mL of leachate, stock solutions of H_2O_2 and $K_2S_2O_8$ were added to the reactor at predetermined doses introduced at doses reaching complete stoichiometric decomposition depending on the COD concentrations (59). After pH adjustment, during UV-PS/H_2O_2 treatment, pre-treated leachate and peroxides were mixed with aeration pump at room temperature. In the Heat-PS/H_2O_2 treatment, they were magnetically stirred and shaken continuously at different predetermined temperatures between 35 and 80 °C, in a 1 L beaker. All experiments were performed in the pH range of 4-11, persulfate dose (1.5, 2.5, 3, and 4 g L^{-1}), H_2O_2 dose (0.5, 1.5, 2, and 3 g L^{-1}), reaction time of 20-90 minutes, and temperature of 35-80 °C for Heat-PS/H_2O_2 AOP. The values and ranges of the independent variables were determined based on previous studies (59-63).

Initially, the aeration tank was inoculated with acclimatized activated sludge prepared from the Behshahr compost leachate treatment plant. Then, by transferring the wastewater from the leachate oxidation stage to the aeration tank, the working volume of the leachate was fixed at 4 L. To set up the system, after feeding the reactor with leachate, aeration was done for about 18 hours. Then, a part of the treated leachate was discharged. The calculated sludge retention time (SRT) was between 12-23 days for different leachate dilutions. The air was uniformly introduced into the leachate at the bottom of the AT through a pump and pipe equipped with a diffuser. The pH of the pre-treated leachate ranged from 6.2 to 8.6. The temperature of the leachate varied from 21 to 25 °C. After reaching a steady state, the leachate treatment efficiency was stabilized, and monitoring of the performance of the EAAS system was conducted. The leachate aerobic treatment process's values and the range of parameters were determined based on preliminary studies and a literature review (64,65). The effluent resulting from different treatment processes by BFLTS is shown in Figure 2.

Statistical Analysis

Data were analyzed using SPSS version 25 and statistical methods of one-way analysis of variance (ANOVA) and

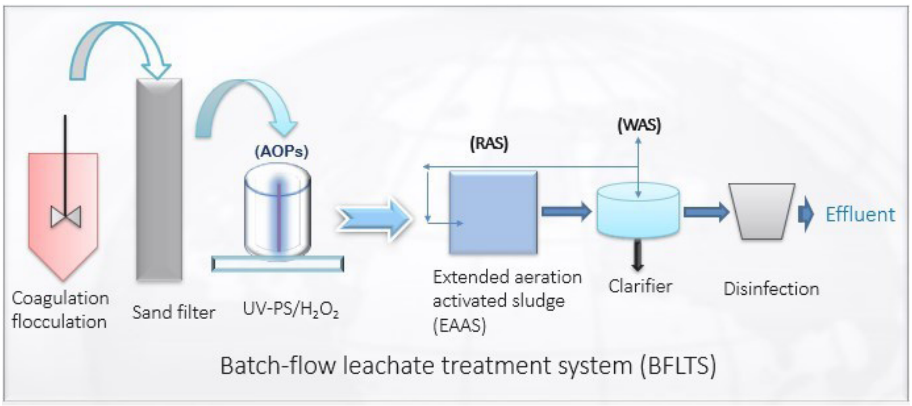

Figure 1. Batch-flow leachate treatment system (BFLTS) image

post hoc tests. When the *P* value is at < 0.05, there is a significant difference between the variables. Measurements were expressed as mean ± standard deviation.

Results

Leachate characteristics

Table 1 shows the characteristics of leachate obtained from the municipal landfill in Qaem-Shahr city. It presents data on the maximum, mean, and minimum values of physicochemical properties, COD/BOD_5 levels, and the actual characteristics of the leachate. The results of this study indicate that the leachate contains significantly high levels of both COD and BOD_5. This suggests that the leachate may have a substantial impact on the receiving water body. Therefore, it is crucial to implement effective treatment measures to mitigate the potential environmental impact of this leachate.

Coagulation/flocculation stage

The study found that $FeCl_3$ was the most efficient coagulant for removing both COD and turbidity from the leachate. When $FeCl_3$ was used, significant reductions in COD (up to 58%) and turbidity (up to 72%) were achieved. In comparison, alum ($Al_2(SO_4)_3$) and $FeSO_4$ only achieved moderate reductions, with COD reductions between 33 and 46% and turbidity reductions between 29% and 35%.

The highest removal of COD and turbidity was achieved at pH 6 using a $FeCl_3$ concentration of 0.8 g L^{-1}. Therefore, $FeCl_3$ was selected as the coagulant for the flocculation-coagulation process.

After the flocculation-coagulation process, the effluent was then filtered through a sand filter to remove any residual floc before undergoing AOPs. This additional filtration step helps ensure that the leachate is free from any remaining particles or impurities before further treatment.

Figure 2. The effluent obtained from various treatment processes by batch flow leachate treatment system (BFLTS)

The values of the removal efficiency (%) of the parameters COD, BOD, TKN, and turbidity for the two different concentrations of leachate samples, maximum (L1), minimum (L2), in the process of coagulation-flocculation using iron chloride are presented in Figure 3.

Advanced oxidation processes

The comparison of the removal efficiency of different AOPs in the COD removal of C-F/SF effluents was done in this study and is shown in Figure 4. Each of the UV-H_2O_2, UV-PS, UV-PS/H_2O_2 and Heat-PS, Heat-H_2O_2 and Heat/PS-H_2O_2 processes was effective in removing pollutants from leachate treatment. In these processes, oxidants are activated by UV radiation or heat. The UV-PS/H_2O_2 process showed the highest level of leachate purification in removing COD, BOD, TKN, and turbidity.

Following the preliminary study, the optimal treatment process and its specific conditions for leachate with the highest COD removal capacity were identified at pH 7, utilizing UV radiation at 15 W (wavelength of 254 nm) with a reaction time of 70 minutes in the UV-PS/H_2O_2 process. The COD removal capacity was found to be 83.5% and 87.4% for leachates with the highest and lowest COD content, respectively ($P < 0.05$). The impact of key parameters (COD, BOD, TKN, turbidity) on two types of leachates (maximum (L1), minimum (L2)) in the AOP process (UV-PS/H_2O_2), employing the optimal amount of oxidant and pH, is depicted in Figure 5.

The COD removal efficiency in UV-activated photocatalytic AOPs decreases when the concentration of H_2O_2 exceeds 1.5 g L^{-1}. Therefore, it was determined that 1.5 g L^{-1} is the optimal amount of H_2O_2. Conversely, all UV-activated processes utilizing 2.5 g L^{-1} persulfate showed enhanced efficiency in COD removal.

By increasing the concentration of PS up to 3 g L^{-1} and H_2O_2 up to 2 g L^{-1} in the heat-activated peroxides process, the rate of COD removal increased. However, it was observed that peroxides are not very effective at low

Table 1. Characteristics of leachate collected from the municipal landfill of Qaem-Shahr city

Parameter	Winter			Summer		
	Maximum	Mean ± SD	Minimum	Maximum	Mean ± SD	Minimum
COD (mg/L)	31620	23900 ± 6303	16180	34260	26737 ± 6142	19214
BOD_5 (mg/L)	12556	7966 ± 3747	3376	16142	10636 ± 4495	5131
BOD_5/COD ratio	0.39	0.29 ± 0.07	0.2	0.47	0.36 ± 0.08	0.26
pH	8.6	7.5 ± 0.85	6.5	8.3	7.3 ± 0.81	6.3
EC (µS/cm) (20°C)	12470	9455 ± 2461	6440	15360	11140 ± 3445	6920
TSS (mg/L)	5640	3814 ± 1490	1989	5500	3450 ± 1673	1400
Turbidity (NTU)	981	679 ± 246	378	1100	725 ± 306	350
TKN (mg/L)	2130	1490 ± 522	850	1950	1280 ± 547	610

temperatures. At 35°C, the reaction rate with the organic matter and the COD removal efficiency by peroxides were both very slow. The highest COD removal efficiency was achieved when using binary and triple processes with a dose of 3 g L^{-1} PS and 2 g L^{-1} H_2O_2, activated at 65°C for a reaction time of 90 minutes and applied at pH 7. Under these optimal conditions, the COD removal efficiency for each process was as follows: Heat-PS/H_2O_2 > Heat-PS > Heat-H_2O_2, with values of 74.8%, 58.6%, and 34.5%, respectively ($P < 0.05$).

In all processes, increasing the initial leachate pH from 4 to 7 increased COD removal efficiency. However, when the pH was raised to 9, the effect on the photocatalytic process was minimal and led to a decrease in efficiency.

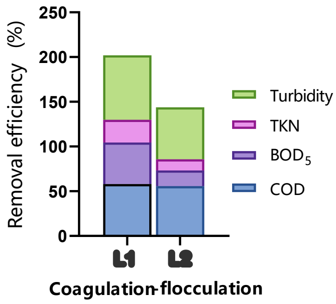

Figure 3. Removal efficiency (%) of four experimental variables of the landfill leachate with coagulation/flocculation pretreatment using $FeCl_3$ (0.8 g L^{-1} and pH 6)

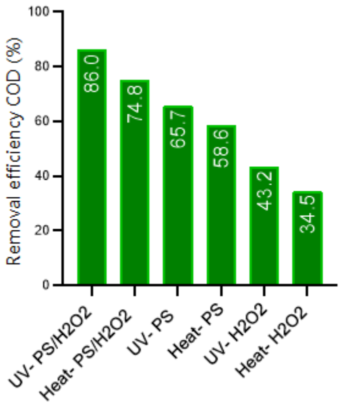

Figure 4. The comparison of the removal efficiency of different advanced oxidation processes in the COD removal of C-F/SF effluents

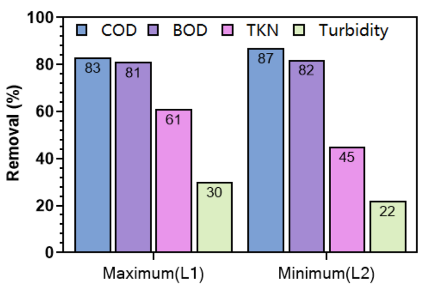

Figure 5. The removal efficiency of four experimental variables of coagulation/flocculation/sand filtration effluent with AOP (sulfate-hydroxyl radical) using UV (2.5 g L^{-1} PS, 1.5 g L^{-1} H_2O_2, pH 7, UV 15 W, temperature = 25 ± 2°C, time = 70 min)

Additionally, in the triple process, increasing the pH from 9 to 11 further intensified the declining trend of COD removal efficiency. The influence of irradiation time and contact time, both important parameters, were investigated and the results are shown in Figure 6.

Extended aeration-activated sludge stage

In this phase, EAAS processes were applied to the treatment of the organic matter in leachate in coagulation/flocculation/sand filtration/AOP effluent, by applying optimal operating conditions for efficient removal of organics using an EAAS reactor. After a three-week adaptation period of activated sludge, the MLSS concentration was maintained in the range of 2900-3500 mg L^{-1} and the COD removal remained constant at 75%. The average BOD_5 and COD of the effluent entering the extended aeration treatment system were 863 and 1546 mg L^{-1}, respectively. In the EAAS process, COD and biological oxygen demand removal rates were 84.2% and 87%, respectively, with hydraulic retention time (HRT) of 18 hours and 87.7 and 91.7% with HRT of 36 hours ($P < 0.05$). The removal efficiency of four experimental variables of leachate under optimal conditions is shown in Table 2.

The overall performance of the BFLTS in MSWL treatment

Table 2 provides a summarized overview of the effect of BFLTS on the efficiency of removing four experimental variables in the treatment process of leachate from Qaem-Shahr city's municipal landfill, under optimal conditions. The BFLTS system demonstrated an average removal performance of 99.2%, 83.2%, and 87.1% for COD, TKN, and turbidity pollutants, respectively.

In this study, the final treated leachate exhibited varying COD effluent concentrations for each type of leachate investigated. The maximum and minimum COD concentrations were found to be 278 ± 25 and 106 ± 10 mg L^{-1}, respectively.

Other physicochemical characteristics of the effluent for maximum and minimum leachates, respectively, include pH 6.4, 7.3, electrical conductivity (EC) 531, 410 µS/cm,

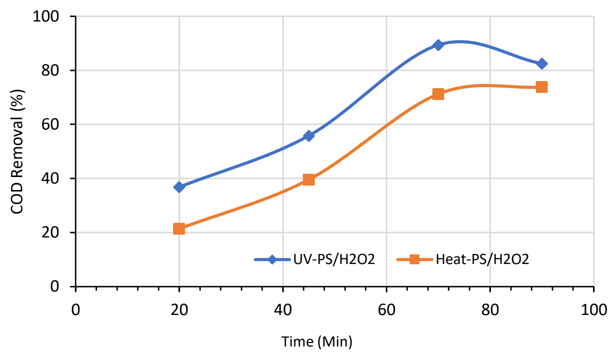

Figure 6. Comparison of leachate COD removal efficiency over time for UV-PS/H_2O_2 and Heat-PS/H_2O_2 processes

Table 2. Impact of BFLTS on the removal efficiency of four experimental variables in the treatment process under optimal conditions for leachate from the municipal landfill of Qaem-Shahr city

Type leachate	Parameter	Raw Leachate	C-F/SF	UV- PS/ H_2O_2	EAAS
Maximum	COD (mg/L) (% removal)	31620	13280 ± 460 (58)	2191 ± 70 (83.5)	278 ± 25 (87.3)
	BOD (mg/L) (% removal)	12556	6730 ± 350 (46.4)	1245 ± 15 (81.5)	99 ± 10 (92)
	BOD_5/COD ratio	0.39	0.5	0.57	0.35
	TKN (mg/L) (% removal)	2130	1586 ± 75 (25.5)	610 ± 20 (61.5)	345 ± 15 (43.4)
	Turbidity (NTU) (% removal)	981	274 ± 10 (72)	191 ± 5 (30.2)	101 ± 3 (46.8)
Minimum	COD (mg/L) (% removal)	16180	7151 ± 300 (55.8)	901 ± 30 (87.4)	106 ± 10 (88.2)
	BOD (mg/L) (% removal)	3376	2785 ± 150 (17.5)	481 ± 15 (82.7)	41 ± 3 (91.4)
	BOD_5/COD ratio	0.2	0.38	0.53	0.38
	TKN (mg/L) (% removal)	850	743 ± 50 (12.5)	407 ± 10 (45.2)	161 ± 10 (60.4)
	Turbidity (NTU) (% removal)	378	158 ± 10 (58)	122 (22.6)	56 ± 2 (53.5)

TSS 169, 138 mg L^{-1}, and the color of the final effluent was clear in all types. In this study, an average of 82, 86, and 58% of metals were successfully removed during C-F/SF, UV-PS/H_2O_2, and EAAS, respectively. The highest removal rate of metals, arsenic and nickel, was 70% and 80%, respectively, in the UV-PS/H_2O_2 process.

In this study, the effluent for the maximum and minimum leachates exhibited different physico-chemical characteristics. The pH values were measured at 6.4 and 7.3, while the EC levels were recorded as 531 and 410 µS/cm for the maximum and minimum leachates, respectively. TSS were found to be 169 and 138 mg/L^{-1} for the maximum and minimum leachates, respectively. The color of the final effluent was clear in both cases.

Discussion

Pretreatment of leachate with the process of coagulation, flocculation, and reduction of hydroxyl and sulfate radical substances absorbents causes better penetration of light in the solution and increases stimulation of the chemical oxidation reaction, and as a result, its effectiveness in leachate treatment increases. The presence of minerals and organic substances in the leachate, pH, stirring velocity, and reaction time are among the factors involved in the efficiency of physicochemical methods in leachate treatment (17,54,66). Consistent with the results of other studies conducted in this field, this research showed that $FeCl_3$ is superior to the other two coagulants examined for the removal of all monitored pollutants in the leachate (57,67). Mainly due to the high concentration of humic substances and organic compounds in the leachate, the color of the leachate is black (57,68). Re-stabilization and lack of proper sedimentation of clots and the effect of leachate color and coagulant have a significant effect on the coagulation effect ($P < 0.05$) (58,69,70). In the treatment of landfill leachate using flocculation-coagulation and optimizing color removal efficiency, polyphenols and nitrates achieved removal efficiencies of 68.8%, 77.5%, and 81.0%, respectively. The optimal conditions in this study were found to be pH 7.66, a coagulant dose of 9.5 g/L, a flocculant dose of 9.1 ml/L, and a stirring time of 10 minutes (71).

During the UV/H_2O_2 process, more active OH radical species are produced by the activation of H_2O_2 by UV rays, which causes further oxidation of organic matter in solid waste leachate (72,73). In the use of AOPs based on hydroxyl and sulfate radicals to remove persistent organic pollutants from wastewater, the radicals react with many organic chemicals at nearly emission-controlled rates. The released radicals are the initiators of the reaction in advanced photocatalytic oxidation processes and can decompose organic materials (74,75). The landfill leachate, treated with 4 UV lamps and 232.7 mM H_2O_2, achieved 72% and 65% removal efficiencies for color and COD in 300 minutes. In contrast, the less concentrated leachate (20% strength) achieved 91% color and 87% COD removal in just 120 minutes. These results demonstrate the effectiveness of the UV/H_2O_2 process as a pre-treatment or treatment technology for landfill leachate (76).

At H_2O_2 concentrations of more than 1.5 g L^{-1}, the removal efficiency of COD decreases by the UV-activated photocatalytic AOPs. Therefore, the concentration of 1.5 g L^{-1} was chosen as the optimal dosage of H_2O_2. All UV-activated processes at 2.5 g L^{-1} persulfate showed higher efficiency in COD removal. This condition might be due to the high reactivity of PS compared to H_2O_2 under catalyzer activation (77). During the elective reaction, the less reactive H_2O_2 may not be completely photolyzed and produce a lower concentration of ion (78). According to the results of other studies, this study also showed that the rate of COD removal with UV/PS as a treatment method was much faster compared to UV/H_2O_2 (79,80). The findings of the study evaluating the effectiveness of UV/Fe^{2+}/H_2O_2 and UV/Fe^{2+}/$S_2O_8^{2-}$ processes in the treatment of landfill leachate pollutants showed that UV/Fe^{2+}/$S_2O_8^{2-}$ had a better performance than Photo-Fenton, and achieved 76.34% COD, 71.44% TOC and 88.94% color removal, compared to 65.58% COD, 48.12% TOC, and 86.65% color removal. Optimizing the oxidant dose and using coagulation/flocculation techniques increased the photocatalytic efficiency. In addition, UV light was observed to have the least effect due to the dark color of the leachate (81).

UV lamp power is one of the effective parameters in photocatalytic processes (82). When exposed to UV rays, peroxides absorb photons, leading to the formation of electron pairs (83). At higher-intensity radiation, peroxides absorb more photons, leading to the formation of more electron pairs. By increasing the leachate turbidity and decreasing the absorption of UV photons, the effect of UV intensity on the activation of peroxides and the removal of organic substances is reduced (84). In addition, the reduction of UV penetration in the leachate occurs in response to the increase in the concentration of organic matter, and thus, prevents the activation of oxidants (85,86). The UV irradiation process alone was ineffective for COD removal (COD removal is only 10%-20%) (87). On the other hand, in the investigation of the effectiveness of PS without the presence of UV radiation in reducing the leachate COD, only 5% COD removal was achieved. Therefore, PS alone is not effective in reducing leachate COD (58). Although the COD removal efficiency decreased with the increase of the peroxide dosage above the optimum level, the COD removal efficiency was lower at the dosages below the optimum level. The increase in the removal rate of COD with the increase in the concentration of peroxides was caused by the production of $SO_4^{\bullet-}$ and $^{\bullet}OH$ radicals in response to the increase in the concentration of PS and H_2O_2. But with the increase of PS concentration from the optimal amount, the $SO_4^{\bullet-}$ radical becomes PS radical with the oxidation-reduction potential lower than the $SO_4^{\bullet-}$ radical (88,89). Also, $SO_4^{\bullet-}$ radical can act as a scavenger radical and become an agent that facilitates the conversion of the $SO_4^{\bullet-}$ radical to PS (90). The synergistic process based on PS/H_2O_2 activated with UV has better efficiency in removing COD from the leachate. The combined $K_2S_2O_8$ and H_2O_2 work better as oxidants if they are added together than if working separately (91).

At higher temperatures, the removal rate decreased, which may be due to the effect of temperature on the rate of H_2O_2 and PS decay (59,92,93). The synergistic effect of oxidants is very effective in leachate treatment. This increase in removal efficiency was achieved in triple processes. Using both H_2O_2 and persulfate reagents, the performance and efficiency of leachate oxidation improved. In the PS/H_2O_2 process under the best conditions, the removal efficiency was 81% and 83% for COD and NH_3-N, respectively (91). In different AOP processes, the removal efficiency of leachate pollutants was higher at pH 7 ± 0.2, and in leachate with higher pH, the pH decreased with time. Previous studies have reported a decrease in pH over time and a neutral effluent when using UV/PS for leachate treatment, which is consistent with the findings of this study (58,94,95). Reducing the pH by reducing the level of alkalinity reduces the degree of inhibition of CO_3^{2-} and HCO_3^- and increases oxidation efficiency (59).

Using biological treatment methods to treat solid waste leachate alone and independently is not very effective due to the high organic load and the presence of toxic substances and heavy metals in the leachate, because it has an inhibitory effect on the growth and metabolism of the microbial mass (96-98). The application of EAAS systems has proven to be successful in treating landfill leachate by achieving significant removal rates. Specifically, these systems have been able to remove COD by 97.03% to 98.87% (99), TN by 81.5% (100), and 77.1% (101).

The standards for discharging treated sewage effluent into the environment typically specify the maximum limits for various parameters, which can vary based on local and national regulations. In Iran, the standards for discharging wastewater and reusing it for surface water, wells, agriculture, and irrigation differ. For instance, the maximum permissible limits for BOD_5, COD, and TSS for discharge into surface water are 30 mg/liter (instantaneous 50), 60 mg/liter (instant 100), and 40 mg/liter (instant 60), respectively, with an opacity limit of 50. The standard pH for discharge into surface water is 6.5-8.5 (102).

When comparing the results of this research with Iranian standards, the close alignment of the values obtained in the effluent of the BFLTS treatment system with the regulatory standards indicates its high efficiency in effectively removing impurities through this combined system. The concentration of the parameters in the effluent is detailed in Table 2.

Treated leachate must be disinfected using processes such as chlorination, UV-radiation treatment, or ozonation before entering the receiving environment and water sources. This is done based on the effluent quality, ease of installation, and ease and cost of maintenance and operation, as well as the effects on plants, animals, and recreational users of the reuse and disposal of the final effluent to the respective receiving waters (103-105).

The biodegradability ratio, which typically decreases over time, is considered a measure of the biodegradability of an organic matter (14). The average BOD_5/COD ratio of effluent in the C-F/SF, UV-PS/H_2O_2, and EAAS systems were 0.44, 0.55, and 0.36, respectively. Therefore, it shows that due to the lower degradability of organic matter in the effluent of the EAAS system, more biological treatment was achieved. Persulfate/H_2O_2 was more effective in increasing the biodegradability of leachate, and during oxidation processes, the ratio of biodegradability increased from 0.09 to 0.17 (91). Based on the data, various types of AOPs have been employed to effectively degrade organic materials under diverse operating conditions. Among these processes, the UV-PS/H_2O_2 AOP has shown promising results and can be considered as a highly efficient treatment method for MSWL treatment. The leachate treatment study showed significant improvement in performance, with a removal efficiency exceeding 80% for all parameters studied, when AOP were combined with biological treatment. This integration enabled compliance with discharge limits, ascribed to the biological removal of biodegradable compounds generated by UV/H_2O_2 treatment (106).

The removal efficiency of three different treatment processes used by a BFLTS was evaluated for four experimental variables. C-F/SF, UV-PS/H_2O_2, and EAAS systems showed average efficiencies of 57%, 85%, and 87% in COD removal and 32%, 82%, and 91% in BOD removal in all types of leachates. UV-PS/H_2O_2 and EAAS processes performed better than conventional C-F/SF processes in removing COD and BOD. The UV-PS/H_2O_2 process demonstrated the highest efficiency and best performance in removing TKN from leachate, achieving an average rate of 53.3%, outperforming other processes. The turbidity removal performance in each of the C-F/SF > EAAS > UV-PS/H_2O_2 processes was 65%, 50%, and 26%, respectively. The results indicated that the UV-PS/H_2O_2 system generated a significantly higher-quality effluent compared to the C-F/SF and EAAS systems in terms of COD and TKN. Additionally, the C-F/SF system outperformed other processes in removing turbidity.

It is crucial to take into account and acknowledge other potential factors that could affect the performance of the methods used to remove or reduce the investigated parameters. These factors may encompass variations in environmental conditions, the presence of co-existing contaminants, and potential interactions between the treatment processes and the specific characteristics of the leachate (37,107,108). By recognizing and examining these additional factors, a more comprehensive understanding of the overall effectiveness of the treatment methods can be attained.

The combined process of coagulation, flocculation, advanced oxidation, and extended aeration in solid waste leachate treatment has several economic aspects (15,24). The combination of coagulation, flocculation, advanced oxidation, and extended aeration processes can lead to improved treatment efficiency and performance (109,110). Coagulation and flocculation help in the removal of suspended solids and organic matter, while AOPs such as UV-PS/H_2O_2 or UV/PS can further degrade recalcitrant organic compounds (17,111). Extended aeration provides additional biological treatment to remove remaining organic pollutants (112). The overall improved treatment performance can lead to reduced treatment costs by minimizing the need for additional treatment steps or reducing the amount of chemicals required for treatment.

The use of coagulants, flocculants, and advanced oxidation agents can be optimized through the combined process (109,113). By effectively removing contaminants in the early stages of treatment, the need for excessive chemical dosing can be minimized. This reduction in chemical usage can lead to cost savings and lower operational expenses. Extended aeration, which involves the use of biological processes to treat organic pollutants, can be more energy-efficient compared to other treatment methods (114,115). The combined treatment process can lead to the minimization of sludge production, particularly through the use of AOPs, which can degrade organic compounds to simpler, more biodegradable forms (110,116). This can result in lower disposal costs and reduced handling and transportation expenses associated with sludge management.

The combined treatment process can ensure that the treated leachate meets stringent environmental regulations and discharge standards (117). By achieving high treatment efficiency and pollutant removal, the facility can avoid potential fines and penalties associated with non-compliance, thus, reducing overall operational costs (118,119). In summary, the integrated approach of coagulation, flocculation, advanced oxidation, and extended aeration in the treatment of solid waste leachate offers significant economic advantages. These benefits stem from enhanced treatment effectiveness, decreased reliance on chemicals, improved energy efficiency, reduced sludge generation, and adherence to environmental regulations. Collectively, these factors contribute to cost savings and overall economic gains for the treatment facility.

Strengths, limitations, and future research

This study investigated the treatment of landfill leachate using a comprehensive approach that includes coagulation, flocculation, advanced oxidation, and extended aeration. The authors emphasize the importance of further research into various advanced chemical oxidation methods and the use of combined treatment systems tailored for leachate treatment, with a focus on natural biological treatment. Due to the complexity and instability of chemical oxidation processes, there are limitations in accurately understanding these processes and providing appropriate scientific solutions. A detailed examination of the removal of resistant and hazardous pollutants from the environment is crucial. These pollutants can have very adverse effects on human health and the environment. The assessment of the efficiency of removing wastewater with higher levels of pollution requires an evaluation of the possibility of integrating other processes. The use of physical processes such as ultrafiltration and nanofiltration can also improve the removal efficiency of resistant and hazardous pollutants from leachate. Ultimately, a thorough examination of the removal of resistant and hazardous pollutants from wastewater requires an evaluation of various treatment processes and the possibility of integrating them. These actions can contribute to enhancing the removal efficiency of pollutants and improving the quality of leachate.

Conclusion

The study findings show that the evaluated treatment processes, including C-F/SF, UV-PS/H_2O_2, and EAAS, showed significant effectiveness in removing pollutants from municipal landfill leachate. The BFLTS system showed strong performance in removing various pollutants such as COD, BOD, TKN, TSS, and turbidity. Overall, this study shows that flocculation-coagulation followed by the AOP process and extended aeration can be a promising and efficient treatment method for landfill leachates. The results of the study highlight the effectiveness of combining physicochemical and biological processes to improve the removal efficiency of pollutants from leachate. This highlights the potential of combined treatment methods in effectively addressing the challenges associated with leachate pollution. The findings suggest that the synergistic approach of employing multiple treatment processes can significantly improve the overall remediation of leachate-contaminated environments.

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Competing interests

The authors declare that there is no conflict of interests.

Ethical issues

This study was approved by the Ethics Committee of Mazandaran University of Medical Sciences (Ethical code: IR.MAZUMS.REC.1401.377). The authors affirm that all the data gathered during the study are accurately presented in the manuscript, and no information from this study has been or will be published independently elsewhere.

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