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The competency of various applied strategies in treating tropical municipal landfill leachate

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ABSTRACT

Leachate is a major pollution source associated with municipal solid waste landfill. This study was carried out to evaluate the effectiveness of various techniques in treating mature landfill leachate generated from landfill in Malaysia, a tropical country. Treatment processes such as biological, ion exchange, coagulation–flocculation, adsorption, advanced oxidation processes (AOPs), and flotation were analyzed. The efficiency of each process was analyzed based on chemical oxygen demand (COD), color, and NH_3-N percentage removals. Ion exchange treatment via cationic/anionic sequence achieved the best removal of color (96.8%), COD (87.9%), and NH_3-N (93.8%) from leachate as compared with other treatment methods. Coagulation–flocculation and AOPs were successful for COD and color removals from leachate. However, both could not treat NH_3-N effectively. Biological treatment could remove NH_3-N (71%) effectively, but it was poor in terms of COD (29%) and color (22%) removals. Adsorption via a new carbon–mineral composite exhibited better removal of both COD (68.4%) and NH_3-N (92.6%) from stabilized leachate.

Keywords: Municipal waste; Landfill leachate; Pollutants; Treatment techniques; Removal efficiency

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1. Introduction

Municipal landfill is one of the most broadly used techniques for the disposal of municipal solid waste (MSW) around the world due to its advantages such as simple disposal procedure, inexpensive, and landscape-restoring effect on holes from mineral workings. Moreover, sanitary landfill is considered as one of the best methods for MSW management all over the world. According to Rafizul and Alamgir [1], a landfill is sanitary, if the landfill design incorporates a leachate collection system to avoid groundwater contamination. In this viewpoint, sanitary landfill will be a safe facility to dispose MSWs which are non-hazardous. Hazardous waste such as chemical, hospital, and radioactive wastes should be carefully managed or should not be allowed to impinge into the municipal landfill. Thus, high standards of environmental protection are a must in the landfill operation [2] which

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includes behavioral guides such as leachate generation and landfill gas emissions [1].

Leachate from MSW landfill is one of the extremely contaminated resources. Bashir et al. [3] and Aziz et al. [4] reported that the generation of highly contaminated leachate can seep into the ground and contaminate the groundwater, surface water, and soil. Fundamental management tool is essential to understand leachate characteristics at a specific site. This is necessary for new polluted landfill and also critical and essential for the old landfill in order to reduce the level of contamination to environment [5]. Landfill leachate contains large amounts of organic compounds (measured as chemical oxygen demand (COD), and color), ammonia, halogenated hydrocarbons suspended solid, significant concentration of heavy metals, and inorganic salts [4,6–8]. El-Fadel et al. [9] acknowledged that the characteristics of leachate can display substantial spatial and temporal variations depending on the site, management practices, refuse characteristics (i.e. age and composition), and internal landfill processes (e.g. hydrolysis, adsorption, speciation, dissolution, ion exchange, redox reactions, precipitation, etc.). Consequently, leachate characteristics may vary from time to time and site to site owing to the variables such as moisture content, waste composition, temperature, and climatic changes [1]. The characteristics of leachate also vary widely with the age of the landfill. Based on its age, landfill leachate can be classified as young leachate (acid-phase, <5 years), intermediate leachate (5–10 years), old or stabilized leachate (methanogenic-phase, >10 years) as illustrated in Table 1 [5]. According to Bashir et al. [7,10] and Halim et al. [8], the most common and serious features of unprocessed stabilized leachate produced from Malaysian landfill sites are its high strength of recalcitrant compounds (as reflected by its COD value) and high level of ammoniacal nitrogen (NH_3-N). The NH_3-N resulting from the decomposition process of organic nitrogen has been recognized not only as a major long-term noxious waste, but also as the primary cause of acute toxicity [11–13]. The existence of high amount of NH_3-N in leachate over a long period of time is one of the most important problems routinely faced by landfill operators [10]. High amount of unprocessed NH_3-N leads to the depletion of dissolved oxygen in surrounding water bodies which is also recognized as eutrophication. NH_3-N concentration of higher than 100 mg/L is very toxic to aquatic organisms [10]. Unless appropriately treated, leachate that seeps from a landfill would contaminate the underlying groundwater.

To choose adequate treatment process which could eliminate contaminates from the leachates, different physicochemical and biological methods or their various combinations could be carried out: (i) biological to remove biodegradable materials [3,14] (ii) ion exchange to remove ammonia and organic compound [10,15], (iii) coagulation–flocculation to remove colloids and metals [16], (iv) adsorption via activated carbon (AC) to remove organics and metals [17], and (v) advanced oxidation process (AOPs) to remove organic compounds [3]. In general, biological processes are not appropriate for the treatment of stabilized leachate, which contains high amount of non-biodegradable organic compounds. Also, the existence of high strength of NH_3-N in the stabilized leachate normally leads to the inhibition of biological activities in the bioreactor [5]. The application of sequencing batch reactor process in landfill leachate treatment achieved optimum removal levels of 25.1, 51.6, and 82.5 for COD, color, and NH_3-N, respectively [15]. As for comparison with physicochemical methods, the application of coagulation–flocculation process in stabilized leachate treatment was reported [16,18]. Amokrane et al. [16] indicated that about 50–65% of COD can be removed effectively from raw stabilized leachate obtained by coagulation–flocculation process. The adsorption of color and COD by a mixture of AC and limestone (15:25 by volume) resulted in 86, 95, 86, and 48% removal efficiencies of color, iron, COD, and NH_3-N, respectively [17]. Less than 15% of COD was removed by air stripping [19]. About 68% of COD and 84% of color were removed from stabilized landfill

Table 1
Landfill leachate classification vs. age [5]

Parameter	Young	Intermediate	Old
Age (years)	<5	5–10	>10
pH	<6.5	6.5–7.5	>7.5
COD (mg/L)	>10,000	4,000–10,000	<4,000
BOD_5/COD	>0.3	0.1–0.3	<0.1
Organic compounds	80% volatile fat acids (VFA)	5–30% VFA + humic and fulvic acids	Humic and fulvic acids
Heavy metals	Low–medium	Low	Low
Biodegradability	Important	Medium	Low

leachate by electrochemical oxidation process [20]. Approximately 50% removal efficiency of COD was achieved by precipitation [21].

Virtually, the published literature merely focused on investigating the efficiency of specific process in treating landfill leachate. Furthermore, review articles simply compared the performances of different biological and physicochemical process in treating landfill leachate collected from different landfill sites throughout the world. In this study, the performances of various applied technologies (i.e. biological, ion exchange, coagulation–flocculation, adsorption, AOPs, and flotation) in treating stabilized landfill leachate generated from a single landfill site were investigated. The main aim of this study was to evaluate the performance of the above-mentioned approaches in terms of organic load and NH_3-N removals from a stabilized leachate generated from a tropical municipal landfill site.

2. Materials and methods

2.1. Characterization of tropical-stabilized landfill leachate

Leachate samples were collected from Pulau Burung Landfill Site (PBLS), at the Byram Forest Reserve in Penang, Malaysia. The PBLS is categorized as a semi-aerobic stabilized landfill and has an area of 62.4 ha, of which 33 ha is currently under operation, receiving approximately 2,200 tons of non-hazardous domestic solid waste daily. The site is equipped with a natural marine clay liner and three leachate collection ponds [13]. Bashir et al. [13] documented that in the first 10 years of operation, from 1980s until 1990, the disposal waste suffered from lack of suitable management and inappropriate leachate control. Starting from 1990, the PBLS is operated as a semi-aerobic system, and the generated leachate is collected through drainage pipes that flows into a collection pond. Generally, a semi-aerobic landfill is an effective method for early stabilization of landfill sites and improvement of waste decomposition. Leachate from a semi-aerobic system is characterized by slightly lower organic matter contents as compared with that in an anaerobic landfill, although still not subjected to biological treatment [22]. The schematic diagram of anaerobic and semi-aerobic (Fukuoka method) landfills is demonstrated in Fig. 1 [22]. The leachate samples were collected manually and placed in 20 L plastic containers. In this study, the samples were taken from the aeration pond due to the effectiveness of the aeration process in eliminating the biodegradable organic matter, resulting in the enhancement of the performance of the treatment processes. The samples were collected at approximately 0.30 m depth as shown in Fig. 2. The samples were immediately transported to the laboratory, characterized, and cooled to 4°C to minimize the biological and chemical reactions. Sample collection and analysis for COD, NH_3-N, color, turbidity, suspended solid (SS), pH, and conductivity were performed in accordance with the Standard Methods for the Examination of Water and Wastewater [23].

2.2. Analytical methods

All tests were conducted in accordance with the Standard Methods for the Examination of Water and Wastewater [23]. Color concentration was measured by Hach DR/2010 spectrophotometer set at 455 nm wavelengths (program number 120) based on the method no. 2120C. Color concentration was reported as platinum–cobalt (Pt–Co/L). The concentration of NH_3-N was measured by Nessler method (Hach method: 8038) using a Hach DR/2010 spectrophotometer at 425 nm wavelength (program number 380). COD concentration was determined by closed reflux colorimetric method (method no. 5220D). COD was measured at a wavelength of 620 nm using a Hach DR/2010 spectrophotometer (program number 435). SS were determined by Hach DR/2010 spectrophotometer set at 810 nm based on Hach Method No. 8006 (program number 630). Turbidity was measured by DR/2010 set at 860 nm according to Method No. 8237. The unit for turbidity is Formazin Attenuation Unit (FAU). pH of the leachate was measured on site, and in the laboratory before and after each test. This was done by a portable digital pH/mV meter (WI-TEG, W-100, Germany). Conductivity was measured as µS/cm by a portable multi-purposes meter (Multi 340i, Germany). All tests were conducted in triplicate in order to obtain consistent results. Removal efficiency of the studied parameters was obtained using the following equation:

$$\text{Removal } (\%) = \frac{C_i - C_f}{C_i} \times 100 \tag{1}$$

where C_i and C_f are the initial and final concentrations of parameters (mg/L), respectively.

2.3. Investigated treatment methods

2.3.1. Biological treatment

This study was conducted to investigate aerobic biodegradation of semi-aerobic stabilized leachate with and without powdered activated carbon (PAC) addition. The leachate contains high levels of NH_3-N with

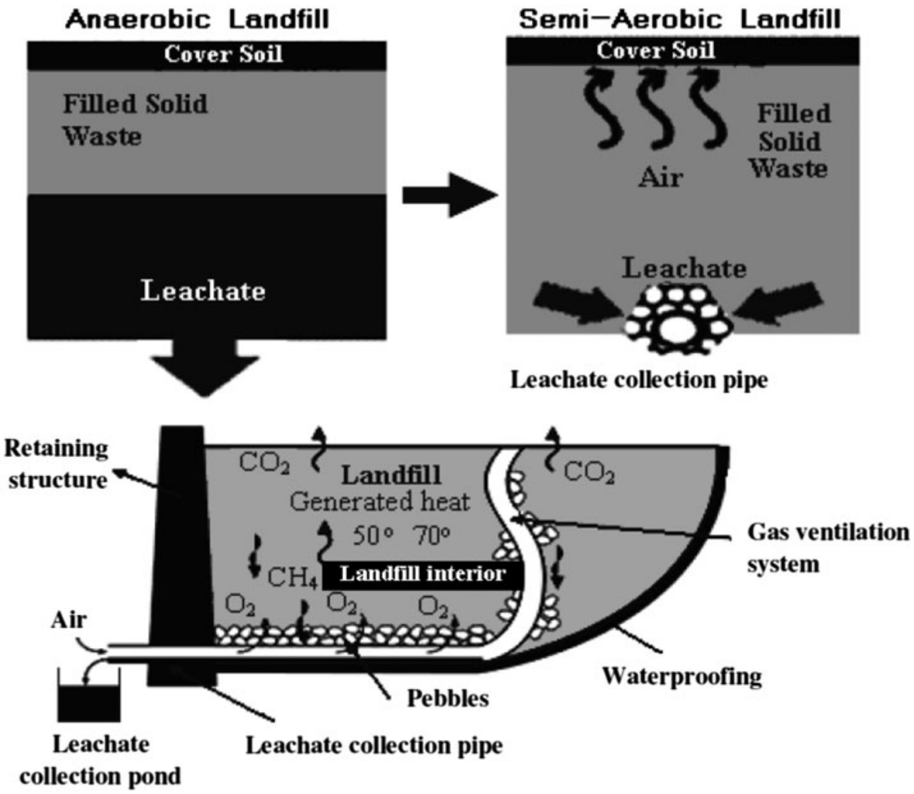

Fig. 1. Schematic diagram of anaerobic and semi-aerobic landfill.

low biodegradability of organic compounds. PAC was added to improve the ability of biological process in treating the leachate [14]. This study involved operating two 16-L semi-aerobic, leachate-fed, laboratory-scale continuous-flow activated sludge (AS) reactors in parallel at room temperature 26 ± 2˚C and pH 6.5 ± 0.5. One reactor was operated without PAC addition (NPAC reactor) while the other one (PAC reactor) was supplemented with PAC of 75–150 μm size to observe its effect on semi-aerobic leachate treatment. The PAC was pre-dried at 103˚C before use. The feed container was located over a magnetic stirrer to keep consistency of feed characteristics. Peristaltic pumps were used to adjust the desired flow rates to the reactors. AS was collected from a textile industry in Penang, Malaysia. The characteristics of AS in (mg/L): total COD 13,800–13,879, soluble COD 224–232, mixed liquor suspended solids 9,740–9,760, mixed liquor volatile suspended solids 8,050–8,070, and sludge volume index 98.5 mL/g, and pH 7–7.66 [24]. In order to check the optimum amount of PAC for the continuous experiments, pre-experiments were firstly conducted [24]. The preliminary experiment was carried out in six 1-L beakers with 600 mL leachate and 200 mL AS. Different amounts (0–4 g/L) of PAC were then added to the beakers and the contents were aerated for two days. Various hydraulics retention times (HRTs), i.e. 0.92, 1.57, and 2.22 d, were also examined. The sludge was acclimated with an initial sludge–leachate mixture of 0.5 L each (50% v/v, leachate-to-activated sludge ratio). The mixture was aerated for one day and 0.5 L of supernatant was withdrawn and replaced by 1L of leachate. Thereafter, the procedure was continued with 0.5 L supernatant withdrawal daily and leachate was fed in increments of 0.5 L, i.e. 1, 1.5, 2, 2.5 L, etc. This was continued till the reactor contents reached 16 L. COD removal and pH were monitored. The acclimatization period was continued for another week. The COD removal was observed to be stable (33%). The acclimated sludge was used as seed in the reactors for further studies [24].

2.3.2. Ion exchange

This study was conducted to investigate the treatability of stabilized landfill leachate using various ion exchange procedures for the first time in literature. A five-stage experiment was conducted to determine the optimum conditions. The optimum operational condi-

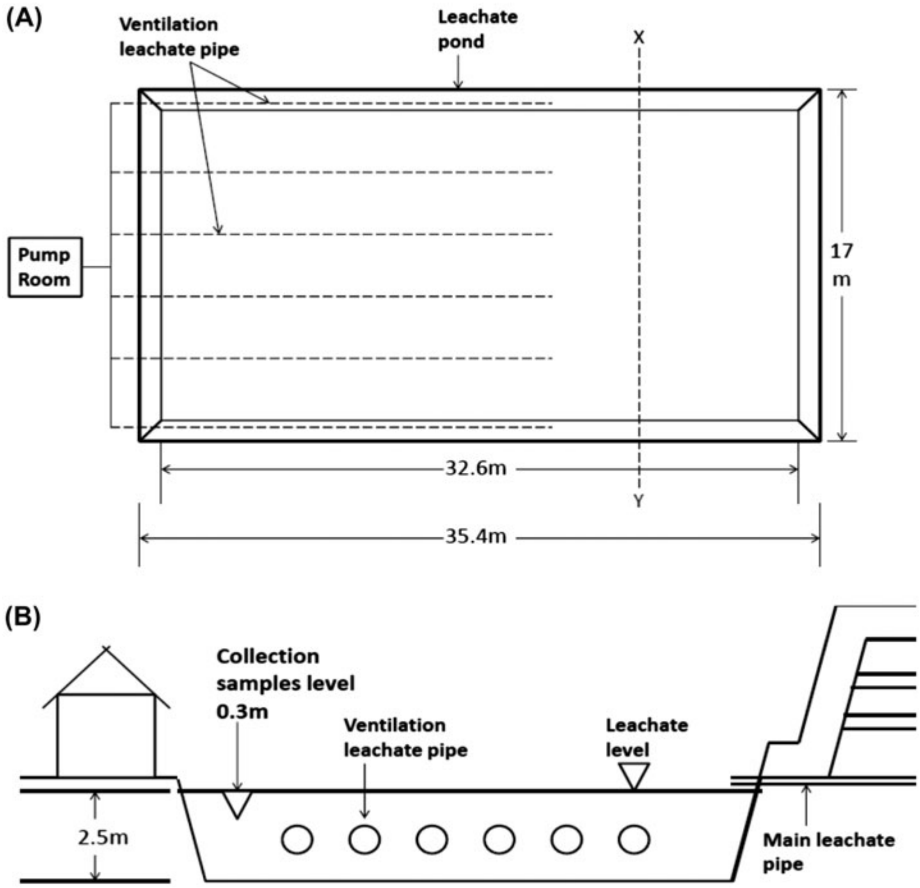

Fig. 2. (A) Illustration of leachate collection pond and (B) vertical cross section ($X - Y$) of leachate collection pond.

tions were selected based on a comprehensive experimental study [25]:

- Stage 1: In this stage, strong acid cationic resin (INDION 225 Na) was utilized for leachate treatment. Different operation conditions were tested such as cation dosage, reaction time, shaking speed, and initial pH.
- Stage 2: In this stage, strong base anionic resin (INDION FFIP MB) was utilized for leachate treatment. Different operation conditions were tested such as anion dosage, reaction time, shaking speed, and initial pH.
- Stage 3: Based on the optimum operational condition obtained from stage 1 and 2, leachate was treated via cationic resin followed by anionic resin. Different dosages of both media were tested to determine the best conditions.
- Stage 4: Based on the optimum operational conditions obtained from stage 1 and 2, leachate was treated via anionic resin followed by cationic resin. Different dosages of both media were tested to determine the best conditions.
- Stage 5: The efficiency of the exhausted media regeneration using various solvents, such as HCl, H_2SO_4, and NaCl, was determined.

Adsorption isotherms are essential for the description of interaction between adsorbent media and pollutants. Therefore, Langmuir and Freundlich isotherm models, empirical equations, were used for the evaluation of experimental results for cation and anion resins.

The Langmuir isotherm is as follows:

$$\frac{1}{q_e} = \frac{1}{QbC_e} + \frac{1}{Q} \quad (2)$$

where C_e is the concentration of adsorbate, (mg/L); q_e is the equilibrium uptake capacity (mg/g); and Q (mg/g) and b (L/mg) are the Langmuir constants. The Freundlich isotherm is expressed as follows:

$$\log q_e = \log K + \frac{1}{n} \log C_e \quad (3)$$

where q_e is the equilibrium uptake capacity (mg/g); K is an indicator of the adsorption capacity in mg/g (L/mg); and $1/n$ is the constant indicator of the intensity of the adsorption.

2.3.3. Coagulation–flocculation

A set of experiments was carried out in order to determine the efficiency of coagulation and flocculation process in treating stabilized leachate using poly-aluminum chloride and aluminum sulfate (alum). The alum used in this study was in powder form with the formula of $Al_2(SO_4)_3 \cdot 18H_2O$ [M = 666.42 g/mol, 51–59% $Al_2(SO_4)_3$, pH 2.5–4] and supplied by Merck, Germany. A hydrolyzed solution of poly-aluminum chloride with the formula of $Al(OH)_xCl_y$ (where x is in the range of 1.35–1.65, and $y = 3 - x$) with the usual acid character (pH 2.3–2.9), due to the presence of hydrochloric acid, was supplied by Idaman Bersih Sdn. Bhd., Malaysia. The 18% solution of poly-aluminum-chloride was used as stock solution throughout the experiments. Coagulation–flocculation was carried out in a conventional jar test apparatus (VELP-Scientifica, model: JLT6, Italy) equipped with six 1-L cylindrical beakers. Stirring was provided mechanically using impellers equipped with 2.5 × 7.5 cm rectangular blades. Operational parameters such as coagulant dosage, pH, speed of rapid mixing (ω_R), duration of rapid mixing (T_R), speed of slow mixing (T_S), and duration of slow mixing (ω_S) were investigated [26].

2.3.4. Adsorption

It is well known that AC is the most commonly used adsorbent worldwide due to its high capability to remove organic compounds from wastewater. However, AC is usually not effective in removing high strength NH_3-N from leachate. Owing to this limitation, the objective of this study was to produce new composite materials for the simultaneous removal of NH_3-N "inorganic compounds" and organic compounds (measured as COD) from stabilized leachate. Thus, a new composite adsorbent material combining admirable properties of AC, zeolite, and low cost adsorbents, i.e. rice husk carbon (RHC) and limestone, was fabricated. Ordinary Portland cement (OPC) was chosen to bind all adsorbents together in a single medium. The study also investigated the capability of the new composite media to remove pollutants from PBLS-stabilized leachate. The process of identifying the optimum composition of the new adsorbent was conducted via batch study. The optimum ratio of hydrophobic (AC to RHC) and hydrophilic media was estimated based on their adsorption properties toward NH_3-N and COD. Batch study was conducted at pH 7, at 5-h contact times, and 250 rpm agitation speed to identify the adsorption properties that produce the optimum ratio. The operational conditions were selected based on the preliminary experiments [27]. The main hydrophobic media were partially replaced by low-cost adsorbent RHC. A three-stage experiment was carried out to determine the optimum conditions.

- Stage 1: The minimum percentage of AC that achieved the highest COD removal was considered the optimum mixture for this media.
- Stage 2: The optimum ratio for hydrophilic media-zeolite (Z) to limestone (L). Zeolite, as the main hydrophilic media, was partially replaced by limestone as a low-cost adsorbent. The optimum mixture was determined at the highest removal of ammonia and at a minimum percentage of zeolite in the mixture.
- Stage 3: The optimum ratio of the combined hydrophobic–hydrophilic media ratio. The optimum ratio of the hydrophilic and hydrophobic media was determined based on the removal patterns of both ammonia and COD. The composite media were tested in batch adsorption study with leachate using both Langmuir and Freundlich isotherm models. The adsorption efficiencies of AC alone, zeolite alone, and the new fabricated media were investigated and reported [27].

2.3.5. Advanced oxidation processes

This part aims to provide an overview about the effectiveness of AOPs employed for PBLS-stabilized landfill leachate treatment. Various AOPs were carried out such as, electrochemical, Fenton, electro-Fenton, persulfate, ozone, ozone/Fenton, and ozone/persulfate oxidation [20,28–30]. For ozone/Fenton and ozone/persulfate processes, both Fenton and persulfate reagents were utilized separately in an ozone reactor as one reaction stage to improve the performance of ozonation for the first time in literatures. A set of experiments was conducted to determine the optimum conditions for AOPs processes including ozone, Fenton and persulfate dosages, pH variation, current density, and reaction time.

2.3.6. Dissolved air floatation + coagulation

This study was conducted to investigate the performance of dissolved air flotation (DAF) for semi-aerobic landfill leachate treatment in batch experiments [31]. Three phases were performed in this work and focused on removing color, COD, and turbidity. The first phase focused on saturator efficiency. The second phase evaluated leachate treatment using DAF alone, while the third phase consisted of coagulation with alum ($Al_2(SO_4)_3$) followed by DAF. Combination of coagulation ($FeCl_3$) and DAF was also investigated to assess the success of these techniques in treating semi-aerobic landfill leachate [32]. Treatment parameters (i.e. flow rate, coagulant dosage, pH, and injection time) were optimized via response surface methodology using central composite design to yield the maximum removal of turbidity, COD, color, and ammonia nitrogen (NH_3-N). Model-determined optimum conditions were tested to confirm the predicted results.

3. Results and discussion

3.1. Leachate characteristics

Stabilized leachate generated from PBLS had a high concentration of COD and high color intensity due to the presence of high molecular weight organic compounds [33,34]. The characteristics of the leachate used in the experiments are summarized in Table 2. The concentration of NH_3-N was also high in PBLS leachates (1,630–2200 mg/L). Low BOD_5 values were observed in PBLS leachate which gave a low biodegradability (BOD_5:COD ratio of < 0.11). Therefore, the current study focuses on removal efficiency of color, COD, and NH_3-N from stabilized landfill leachate. Due to its characteristics, PBLS leachate is recognized as highly stabilized leachate with low biodegradability, hence requiring the application of physicochemical processes for effective treatment. Moreover, low concentrations of heavy metals were observed in PBLS leachate by Aziz et al. [34]. Aziz et al. [34] reported concentrations of Zinc (0.1–1.8 mg/L), Copper (0.1–0.4 mg/L), Manganese (0.6–1.1 mg/L), Cadmium (<0.4 mg/L), and Iron (0.32–7.5 mg/L) in PBLS leachate. Higher values were recorded by other researchers, especially for Iron and Zinc concentration [5,35]. However, the observed decreases of heavy metal concentrations are due to the age and stabilization of landfill [4]. The characteristics also demonstrated that the amount of contaminants exceeded the discharge limits as stipulated by the Malaysian Environmental Quality Act 1974 (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009 [36].

3.2. Investigated treatment methods' performance

3.2.1 Biological treatment

The optimum removal efficiencies for biological treatment with and without PAC for color, COD, and NH_3-N under optimum operational conditions are illustrated in Table 3. It was observed that using the biological treatment, a stable COD removal efficiency of 46% could be achieved at the highest HRT of 2.22 d, corresponding to COD of 2,850 mg/L with the PAC reactor. Whereas for the NPAC reactor under the same conditions, only 29% COD removal was achieved. As the HRT and initial COD concentration were increased, the effect on COD removal efficiency was improved. It was found that at the same HRT of 1.57 d, greater COD removal efficiency (35%) was achieved with the PAC reactor as compared with the NPAC reactor (4.5%). This might be attributed to either possible inhibitory effect of the leachate constituents or COD contribution of the recalcitrant organic compounds which may not have degraded under the experimental conditions the NPAC reactor. The trend of changes in color removal efficiency was quite different for both reactors. The maximum color removal efficiency (31%) was achieved at the highest HRT (2.22 d) for the PAC reactor; whereas, it was only 22%

Table 2
Characteristics of raw leachate from PBLS

Parameter	Units	Measurements Values	Average	Discharge limit
pH	–	8.30–9.17	8.58	6.0–9.0
COD	mg/L	1810–2,850	2,321	400
NH_3-N	mg/L	1,630–2,200	1949	5
Color	Pt–Co	4,250–5,760	5,094	100
Turbidity	FAU	128–330	211	–
SS	mg/L	114–360	181	50
Conductivity	μS/cm	21,850–26,230	24,340	–
Zinc (Zn)	mg/L	0.02–2.0	0.5	1
Total iron	mg/L	0.9–8.8	3.4	5

Table 3
PBLS-stabilized leachate treatment efficiency using aerobic biodegradation with and without PAC

				Removal efficiency, %			Final concentration		
Reactor	Operation conditions	HRT, day	Final pH	COD	Color	NH_3-N	COD, mg/L	Color, Pt.Co/L	NH_3-N, mg/L
PAC	Initial COD, 2,850 mg/L; PAC, 3 g/L	2.22	5.7	46	31	78	1,540	2,895	308
		1.57	7.68	35	12	49	1850	3,695	715
		0.92	6.4	30.7	24	56	1975	3,190	615
NPAC	Initial COD, 2,850 mg/L; without PAC	2.22	7.5	29.2	22	71	2015	3,275	405
		1.57	8.1	4.5	10	40	2,720	3,780	840
		0.92	8.11	19	15	43	2,310	3,570	798

for the NPAC reactor at highest HRT (2.22 d) and highest COD (2,850 mg/L). The low level of color removal with the NPAC reactor was probably due to inhibition resulting from high concentration of leachate in the system and color adsorption by AC in the PAC reactor. In the NPAC reactor, the minimum value of ammonia removal (40%) was obtained at HRT of 1.57 d. The reading rose to a maximum value of 71% when HRT was 2.22 d. The removal efficiencies of COD, color, and NH_3-N were higher in PAC reactor as compared with the NPAC reactor. This indicates that HRT has a greater impact in the removal efficiency [16]. Nevertheless, the results shown in Table 3 indicate that both reactors could not meet the standard discharge limit.

3.2.2. Ion exchange

The treatment efficiencies of the cation resin alone, anion resin alone, cation–anion resin, and anion–cation resin are summarized in Table 4. Optimization of process variables in the case of cation exchange resin alone resulted in removing 68.9, 38.0, and 91.8% of color, COD, and NH_3-N, respectively, under optimized operational conditions (cation dosage, 24.0 cm^3; contact time, 10 min; and shaking speed, 150 rpm). Table 5 shows Langmuir and Freundlich isotherm constants and correlation coefficients. The experimental data for NH_3-N adsorption on cationic resin were compatible with both Langmuir and Freundlich models with $R^2 >$ 0.93. However, the experimental data for color and COD adsorption on cationic resin were incompatible ($R^2 < 0.8$). In case of applying anion exchange resin alone, the optimized conditions (with pH adjustment) occurred at anionic dosage of 35.0 cm^3, contact time of 74 min, shaking speed of 150 rpm, and pH of 3.3. These conditions resulted in 91.7, 70.7, and 11.8% removal of color, COD, and NH_3-N, respectively. However, for the samples without pH adjustment, another optimum operational condition can be selected and applied as following: anionic dosage of 34.4 cm^3, contact time of 74 min, and shaking speed of 150 rpm at pH 8.3. This process yields about 67.1, 61.2, and 15.8% removal of color, COD, and NH_3-N, respectively. Equilibrium removal data of color and COD by anion exchanger fitted well with Langmuir and Freundlich linear adsorption isotherm ($R^2 > 0.96$). The experimental data obtained from both models for NH_3-N adsorption on anion resin were incompatible ($R^2 < 0.44$). Furthermore, the regeneration results indicated that both cation and anion resins could be reused several times with recovery efficiencies of more than 80%. The best performances were obtained when anionic resin and cationic resin were regenerated using 0.2 M of NaCl and 0.3 M of H_2SO_4, respectively.

Ion exchange resins are insoluble materials that contain exchangeable mobile ions. Once the resin comes in contact with the solution, the ion separates and becomes mobile. The ions on the exchanger can be replaced via dissolved ions in the aqueous phase to keep the overall charge neutral [37]. Consequently, removal of positive ions, such as NH_3-N, principally occurs due to the strong exchangeability with the cationic resin mobile ion (H^+) [37]. The interactions between the organic substances in stabilized leachate and anionic resin carrying negative mobile ions is complicated. When an ion exchanger sorbs an organic ion, the hydrocarbon radical of the ion can be engaged in hydrophobic interactions with the matrix of exchangers. As a result, hydrophobic interaction plays a vital role in offering high ion exchange selectivity in favor of aromatic and other hydrophobic ions [38]. The process mechanism of attraction between the exchanger hydrophobic matrix and hydrophobic parts of sorbed ions was discussed by Li and SenGupta [38].

Although the results indicated that anion resin alone could be a valuable and effective alternative for color and COD reductions from semi-aerobic stabilized landfill leachate, the COD values of the final

Table 4
PBLS-stabilized leachate treatment efficiency using ion exchange processes

			Removal efficiency, %			Final concentration		
Media	Operation conditions	pH	COD	Color	NH_3-N	COD, mg/L	Color Pt.Co/L	NH_3-N, mg/L
Cation resin	Sample 100 mL; pH8.3; cation dosage 24.0 cm^3; contact time 10 min; and shaking speed 150 rpm	2.87	38	68.9	91.8	1,440	1,530	160
Anion resin	Sample 100 mL; pH8.3; anion dosage 34.4 cm^3; contact time 74 min; shaking speed 150 rpm	8.05	61.2	67.1	15.08	900	1,680	1,655
Cation–anion	Sample 100 mL; pH8.3; cation dosage of 23.3 cm^3 followed by anion dosage 25.3 cm^3	2.93	87.9	96.8	93.8	280	165	125
Anion–cation	Sample 100 mL; pH8.3; anion dosage 28.3 cm^3 followed by cation dosage of 19.6 cm^3	2.08	72.3	91.6	92.5	640	430	146

Table 5
Langmuir and Freundlich isotherm constants and correlation coefficients

		Langmuir isotherm coefficients			Freundlich isotherm coefficients		
Resin type	Parameter	Q (mg/g)	b (L/mg)	R^2	K (mg/g (L/mg))	$1/n$	R^2
Cation	Color	6.9	0.0022	0.792	0.684	0.313	0.358
	COD	1.11	0.00128	0.753	0.106	0.462	0.574
	NH_3-N	12.66	0.0042	0.984	0.636	0.402	0.935
Anion	Color	15.2	0.00024	0.995	2.128×10^{-6}	2.076	0.979
	COD	3.74	0.00062	0.974	1.87×10^{-8}	2.851	0.951
	NH_3-N	0.406	0.00037	0.477	2.28×10^{-5}	1.378	0.455

effluent were incidentally above the limits allowed by Malaysian laws. In addition, anion resin was inadequate for NH_3-N removal. The optimum effectiveness of sequential treatment applications (cationic–anionic and anionic–cationic) was also investigated. The equations of color, COD, and NH_3-N removals by both cationic–anionic and anionic–cationic treatment sequences were determined. For cationic–anionic treatment, the experimentally achieved optimum removal values of color, COD, and NH_3-N were 96.8, 87.9, and 93.8%, respectively. However, the application of the anionic–cationic treatment resulted in 91.6, 72.3, and 92.5% removal of color, COD and NH_3-N, respectively. Consequently, the results imply that the application of the cationic–anionic sequence for the treatment of semi-aerobic stabilized landfill leachate was more effective than the anionic–cationic sequence. Thus, the cationic–anionic sequence is recommended for the treatment of stabilized landfill leachate.

3.2.3. Coagulation–flocculation

Optimum coagulant dose and pH were, respectively, found at 1.9 g/L and 7.5 for poly-aluminum chloride, and 9.4 g/L and 7 for alum. As shown in Table 6, COD removal of 84.50 and 56.76% was achieved by alum and poly-aluminum chloride, respectively. Using poly-aluminum chloride, almost complete removals for physical parameters of leachate (turbidity: 99.18%, color: 97.26%, and TSS: 99.22%) were achieved; whereas, alum showed inferior removal (turbidity 94.82%, color 92.23%, and TSS 95.92%). Nevertheless, results revealed that poly-aluminum chloride is not as efficient as alum for COD elimination, where the alum dose (9.4 g/L) was about fivefold that of poly-aluminum chloride (1.9 g/L). Based on the above-mentioned results, the performance of coagulation–flocculation process in stabilized leachate treatment can be considered reasonable. Nevertheless, typically, there are sev-

Table 6
PBLS-stabilized leachate treatment efficiency using coagulation–flocculation process

		Removal efficiency (%)				Final concentration		
Coagulant	Operation conditions	COD	Color	Turbidity	NH_3-N	COD, mg/L	Color, Pt.Co/L	Turbidity, FAU /L
Poly-aluminum chloride	Poly-aluminum chloride dosage, 1.9 g/L; pH; 7.5; ω_R, 80 rpm; T_R, 1 min; ω_S, 30 rpm; T_S, 15 min	56.76	97.26	99.18	NA[a]	835	110	2
Aluminum sulfate (alum)	Alum dosage, 9.4 g/L; pH, 7; ω_R, 80 rpm; T_R, 1 min; ω_S, 30 rpm; T_S, 15 min	84.50	92.23	94.8	NA	300	305	13

[a] Not available.

eral drawbacks of coagulation–flocculation process including not effective for NH_3-N removal, lower removal efficiency for high-strength leachate, and production a considerable amount of sludge. Besides that, an increase in the concentration of aluminum in the liquid phase may be observed [17].

3.2.4. Adsorption

The final composition of the proposed composite media for optimum leachate treatment consisted of 45% zeolite, 15.31% limestone, 4.38% AC, 4.38% RHC as adsorbents, and 30% OPC, used as binder. Both the Langmuir and Freundlich isotherm studies indicated that these media show favorable adsorption. Based on the Langmuir model, the adsorption capacities of NH_3-N, COD, and color, reached 24.3, 22.99, and 43.67%, respectively. The composite media exhibited excellent combination adsorption properties toward organic (COD) and inorganic contaminants (such as NH_3-N) in the leachate. As illustrated in Table 7, comparison study indicated that the adsorption capacity of composite adsorbent towards NH_3-N (24.34 mg/g) was higher than zeolite (17.45 mg/g) and AC (6.079 mg/g) and comparable to AC for COD. Fig. 3 illustrates the comparison for removal of COD and NH_3-N which indicated that composite media was an efficient method than other adsorbents.

3.2.5. Advanced oxidation processes

The performance and efficiency of various AOPs in stabilized leachate treatment are summarized and presented in this section. Electrochemical oxidation using graphite carbon electrode obtained optimal removal efficiencies of 68 and 84% for COD and color, respectively. However, the performance of electrochemical oxidation dwindled when aluminum electrode was used with an optimum removal of 49.33 and 59.24% for COD and color, respectively. For Fenton oxidation, the experimental process achieved optimum removal

Table 7
Langmuir and Freundlich isotherm constants and correlation coefficients for the new composite media

		Langmuir isotherm coefficients			Freundlich isotherm coefficients		
Adsorbents	Parameter	Q (mg/g)	b (L/mg)	R^2	K mg/g (L/mg)	$1/n$	R^2
Composite media	Color	43.67	0.02	0.97	8.8838	0.218	0.87
	COD	22.99	9.4×10^{-4}	0.965	0.0642	0.7637	0.9291
	NH_3-N	24.3	2.15×10^{-4}	0.971	0.0188	0.8293	0.9736
Activated carbon	Color	–	–	–	–	–	–
	COD	37.88	1.0×10^{-3}	0.982	0.6875	0.1543	0.9384
	NH_3-N	6.079	1.51×10^{-3}	0.937	0.8866	0.0017	0.9335
Zeolite	Color	–	–	–	–	–	–
	COD	2.3458	1.14×10^{-3}	0.817	1.27×10^{-5}	1.996	0.8051
	NH_3-N	17.4520	3.14×10^{-4}	0.968	0.8287	0.1054	0.9631

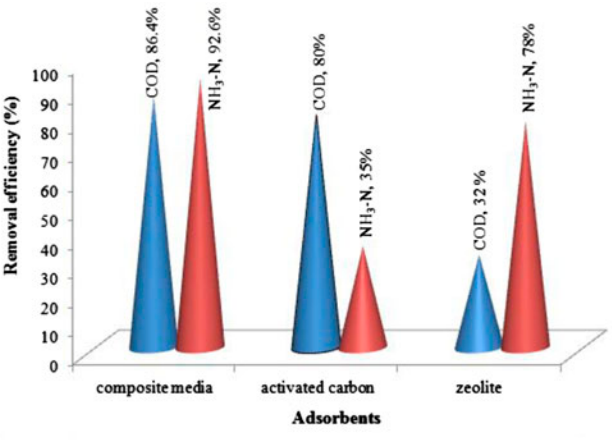

Fig. 3. PBLS stabilized leachate treatment efficiency using a new adsorbent composite media, activated carbon, and zeolite.

values for COD and color of 58.3 and 79%, respectively. However, by electro-Fenton oxidation, the optimum removal efficiency was improved to 94.4 and 96.6% for COD and color, respectively, which revealed that electro-Fenton process is more efficient than both Fenton and electrochemical oxidation. Although persulfate is a newest advanced oxidant reagent used for treatment of different types of wastewaters, the performance of persulfate as sodium persulfate ($Na_2S_2O_8$) in treating stabilized leachate was lower than Fenton oxidation (Table 8).

Optimization process variables of ozone alone achieved 24.7, 90.8, and 6.4% removal efficiencies of COD, color, and NH_3-N, respectively, under the operational conditions of 70 g/m^3 ozone dosage, 60 min reaction time, and 250 mg/L initial COD concentration at natural leachate pH (8.5). However, the removal efficiency becomes lower when higher initial COD concentration was used, as shown in Table 8. Although the removal efficiency was only improved for color as well as slightly improved in COD removal at low leachate concentration. The performance of ozone alone in stabilized leachate treatment is still low particularly in organics and ammonia removals, which suggest utilizing advanced oxidant materials to improve the removal efficiency.

In the case of ozone/Fenton oxidation, the optimization process achieved 78, 98, and 20% removal of COD, color and NH_3-N, respectively, at optimal operational condition of 30 g/m^3 ozone dosage, H_2O_2 0.01 mol/L, Fe(II) 0.012 mol/L dosage, pH 5, and 90 min of reaction time. Consequently, the effectiveness of

Table 8
PBLS-stabilized leachate treatment efficiency using various AOPs

Method	peration conditions	Final pH	Removal efficiency (%)			Final concentration		
			COD	Color	NH_3-N	COD, mg/L	Color, Pt.Co/L	NH_3-N, mg/L
Electrochemical oxidation (graphite carbon electrode)	Na_2SO_4, 1 g/L; reaction time (RT), 4 h; and (CD), 79.9 mA/cm^2	9.1	68	84	NA	600	475	NA
Electrochemical oxidation (Al)	NaCl, 2 g/L; RT, 218 min; and CD, 75 mA/cm^2	NA	49.33	59.24	NA	1,315	1,330	NA
Fenton oxidation	H_2O_2, 0.033 mol/L; Fe(II), 0.011 mol/L; pH, 3; and RT, 145 min	NA	58.3	79	NA	985	885	NA
Electro-Fenton oxidation	RT, 45 min; pH, 3.5; H_2O_2, 0.012 mol/L; Fe^{2+}, 0.012 mol/L; and CD, 55 mA/cm^2	NA	94.4	96.9	NA	135	130	NA
Persulfate oxidation (as $Na_2S_2O_8$)	RT, 240 min; pH: 8.5, COD/$S_2O_8^{2-}$ ratio, (1 g/7 g); rotation, 350 rpm; and Temp. 28˚C	8.35	39	55	22	1,235	1,600	22
Ozone	RT, 60 min; pH, 8.5; and O_3 dosage 80 g/m^3	8.4	15	27	0.25	1,720	2,590	808
Ozone/Fenton	RT, 90 min; O_3 dosage, 30 g/m^3; Ph, 5; H_2O_2 0.01 mol/L; and Fe^{2+}, 0.02 mol/L	3.1	78.0	99.0	20.0	445	35	648
Ozone/Persulfate	RT, 210 min; O_3 dosage 30 g/m^3; pH, 10; and persulfate dosage 1 g/1 g (COD/$S_2O_8^{2-}$ ratio)	9.93	72	96.0	76.0	570	140	195

ozone/persulfate oxidation resulted in 72, 96, and 76% removal efficiencies of COD, color, and NH_3-N, respectively, at optimal conditions of 30 g/m^3 ozone dosage, 210 min of reaction time and persulfate dosage of 1 g/1 g (COD/$S_2O_8^{2-}$ ratio). The obtained results from the comparative experiments revealed that the combination of Fenton and persulfate and advanced oxidant reagents for ozonation improved the treatability of the stabilized leachate. Furthermore, electro-Fenton and ozone/Fenton oxidation processes are much more effective in removing COD and color as compared with other treatment processes; whereas, ozone/persulfate is more effective in removing NH_3-N. The final effluent concentrations of ozone/Fenton processwere as following; COD (392 mg/L) and color (60 Pt.Co.). The readings successfully meet the acceptable discharge level (COD 400 mg/L, color 100 Pt. Co.) as prescribed by the Environmental Malaysian regulation 2009 for solid waste landfill control. Although the removal of NH_3-N by using ozone/persulfate process was much higher than other process; however, the final effluent concentration of NH_3-N (198 mg/L) does not meet the maximum acceptable level (5 mg/L).

3.2.6. Dissolved air floatation + coagulation

In this section, the performances of dissolved air floatation and combination of coagulation processes in semi-aerobic stabilized leachate treatment were investigated and the results are demonstrated in Fig. 4 The best removal in the DAF system combined with alum coagulant was obtained with 4 min injection time, 20 min retention time, and 2.3 g/L alum dos; resulting in 70, 79, and 42% removals of color, COD, and turbidity, respectively. For combined coagulant ($FeCl_3$) and DAF system, the optimum removal values for turbidity, COD, color, and NH_3-N were 50, 75, 93, and 41%, respectively. The optimum operating conditions for coagulation and DAF were 599.22 mg/L of $FeCl_3$ at pH 4.76, followed with saturator pressure of 600 kPa, flow rate of 6 L/min, and injection time of 101s. Although the removal of COD was slightly higher used alum in DAF system, the results revealed that utilizing $FeCl_3$ with DAF was more efficient in semi-aerobic stabilized leachate treatment.

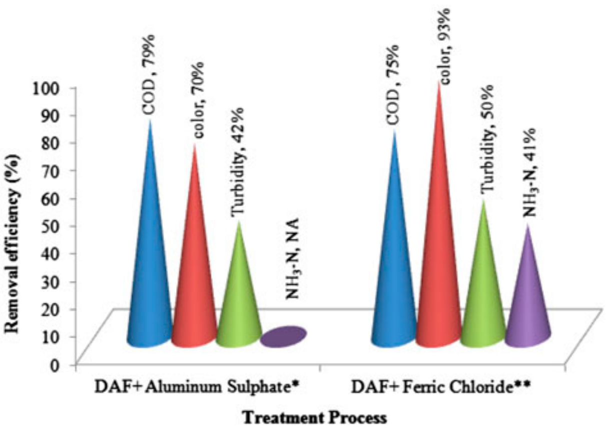

Fig. 4. PBLS stabilized leachate treatment efficiency using DAF + coagulation.

*Process 1 [DAF + coagulation alum ($Al_2(SO_4)_3$)]: injection time, 4 min; alum dose, 2.3; saturator pressure 600 kPa; retention time, g/L 20 min; and flow rate, 6 L/min.

**Process 2 [DAF + coagulation ($FeCl_3$)]: injection time 1 min 41sec; $FeCl_3$ dosage 0.599 g/L; saturator pressure 600 kPa; and flow rate, 6 L/min.

3.3. Performance and limitations of applied technology

The published literature focused on investigating the efficiency of specific process in treating landfill leachate. Furthermore, review articles compared the performance of different biological and physico-chemical process in treating landfill leachate, which were collected from different landfill site in different countries. Nevertheless, in this study, the performance of various applied technologies (i.e. biological, ion exchange, coagulation–flocculation, adsorption, and AOPs) in treating stabilized landfill leachate generated from unchanged landfill site was investigated. Based on the above affirmation results, stabilized leachate treatment efficiency using aerobic biodegradation with and without PAC was in adequate in treating stabilized leachate particularly in removing color and COD.

Applying cation resin alone showed great efficiency for NH_3-N removal (91.8%). But, it showed insufficient removal of color (68%) and COD (38%). Anion resin alone could be an effective alternative for color (67%) and COD (68%) reduction. However, the COD values of the final effluent were incidentally more than the limits allowed by Malaysian laws. Also, anion resin was inadequate for NH_3-N removal. The application of cationic–anionic sequence for the treatment of stabilized landfill leachate was more effective than the anionic–cationic sequence. For cationic–anionic treatment, the experimentally achieved optimum removal values of color, COD, and NH_3-N were 96.8, 87.9, and 93.8%, respectively. Nevertheless, using ion exchange in treating raw leachate is costly due to the high amount of anion and cation resin dosages required for treating a small amount of leachate. Therefore, ion exchange is recommended as a post-treatment process.

Using poly-aluminum chloride is not as efficient as alum for COD elimination, where the alum dose (9.4 g/L) was about fivefold that of poly-aluminum chloride (1.9 g/L). However, typically, there are several drawbacks of this method including not effective for ammonia–nitrogen removal, lower removal efficiency for high-strength leachate, and production of a considerable amount of sludge. Similarly, applying flotation process combined with coagulation resulted in poor efficiency in terms of ammonia removal.

Applying AC alone showed good removal efficiency for COD and color removal, but inefficient in removing ammonia. Zeolite showed better ammonia removal efficiency and poor performance in organics reduction. In comparison with other adsorbents, the performance of the new composite media that combined both AC and Zeolite displayed an excellent performance in removing color, COD, and ammonia from stabilized leachate. However, the method has numerous of drawbacks including the requirement of high amount of media. To bring it further, the residual amounts of pollutants were incidentally above the limits allowed by Malaysian laws (100 Pt–Co/L for color and 400 mg/L for COD) as stipulated by the Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009 under the Laws of Malaysia—that is, the Malaysia Environmental Quality Act 1974. Employing AOPs, such as electrochemical oxidation, Fenton oxidation, and electro-Fenton oxidation, displayed good reduction in color and COD However, their performance in terms of ammonia removal was incompetent. The efficiency of O_3 alone is insufficient for the removal of COD, color, and ammonia (15, 27, and 0.25%, respectively). Persulfate oxidation is more efficient for leachate treatment than O_3 alone. Although the performance of O_3 after $S_2O_8^{2-}$ is improved, the removal efficiency is also improved by the advanced oxidation system ($O_3/S_2O_8^{2-}$) to 72, 93, and 55% for COD, color, and ammonia, respectively, under optimal conditions. The ozone/Fenton in AOPs obtained 65, 98, and 22% removal efficiencies for COD, color, and NH_3-N, respectively.

With reference to the above-mentioned subject, a preliminary treatment including advanced sludge process (ASP), coagulation–flocculation, Fenton oxidation (FO), and electrochemical oxidation, is recommended to be used in treating stabilized landfill leachate followed by ion exchange process which can remove NH_3-N effectively.

4. Conclusions

The capabilities of various treatment techniques for the treatment of old landfill leachate generated from PBLS, Malaysia, were studied. Using electro-Fenton or ozone/persulfate as preliminary treatment is highly recommended for this kind of leachate. It can be concluded that coagulation flocculation process was not efficient in treating ammonia. On the contrary, adsorption via AC, flotation, electrochemical oxidation, Fenton oxidation, and ozonation were not adequate to treat ammonia. Furthermore, the above-mentioned processes were not capable to completely remove color and COD. Thus, further step is required to remove the trace amount of NH_3-N. Using a new composite media or a combination of both anionic and cationic resin resulted in a good quality removal of color, COD as well as NH_3-N. However, the required dosages of media were relatively high, leading to the increase of treatment cost. Also, the exhausted media will create another environmental predicament.

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