

# Sustainable treatment of landfill leachate

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**Abstract** Landfill leachate is a complex liquid that contains excessive concentrations of biodegradable and non-biodegradable products including organic matter, phenols, ammonia nitrogen, phosphate, heavy metals, and sulfide. If not properly treated and safely disposed, landfill leachate could be an impending source to surface and ground water contamination as it may percolate throughout soils and subsoils, causing adverse impacts to receiving waters. Lately, various types of treatment methods have been proposed to alleviate the risks of untreated leachate. However, some of the available techniques remain complicated, expensive and generally require definite adaptation during process. In this article, a review of literature reported from 2008 to 2012 on sustainable landfill leachate treatment technologies is discussed which includes biological and physical–chemical techniques, respectively.

**Keywords** Landfill leachate treatment · Sustainable treatment · Biological treatment · Physical–chemical treatment

## Introduction

The exponential generation of municipal solid waste (MSW) over the years has been contributed mainly due to the expanding of industrial activities, population growth,

and lifestyle changes (Ahmed and Lan 2012). In Malaysia alone, population has been increasing at a rate of 2.4 % every year and the generation of MSW also increases dramatically. As a result, various types of MSWs including industrial, commercial and agricultural byproducts are being disposed to the landfill over the years. Therefore, it is undoubtedly that appropriate MSW management is somewhat crucial (Akinbile et al. 2012) nowadays. Most significantly, Malaysians are currently generating about 5,781,600 tonnes of solid waste annually based on 2012 census data. Put together the waste generation of 0.9 kg/capita/day, it is expected that the amount of solid waste will be increased to double digits as the country is moving forward to be a developed nation in 2020. This estimation is by some means realistic because the process of urbanization has seen many rural and isolated areas receive widespread economic development program which has changed Malaysia landscape entirely due to the implementation of Government Transformation Program (GTP) introduced by the present 6th Malaysia's Prime Minister in 2009.

Consequently, responsible authorities particularly municipalities and landfill operators nationwide are facing difficulty in dealing with staggering amount of MSW to dispose it in a sustainable way. In addition, the selection for ideal and feasible method in controlling the disposal of high quantities of MSW at economical costs that can avoid environmental damages are difficult to be decided due to various deliberations need to be made (Umar et al. 2010). Conventionally, landfilling of solid waste has been the most preferred method for solid waste disposal due to technical feasibility, ease of operation, minimum supervisions and low operation expenditure. In most countries, landfilling is the most acceptable means for eliminating MSW which favors to the technology exploitation and

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capital cost (Renou et al. 2008). While most of the landfills nowadays equipped with a level three sanitary systems, many developing countries are still struggling to equip state of the art facilities at the landfill. For example, there are 261 landfills in Malaysia whereby more than 80 % of them are being controlled tipping or open dumping practice. This is due to the fact that it obscures lower cost of operation and maintenance compared to the other established techniques (incineration and advanced landfill system) (Halim et al. 2010a). Unfortunately, this practice has caused excessive generation of leachate whereby if it is not treated and safely disposed, landfill leachate could be a potential source of surface and ground water contamination, as it may percolate through soils and subsoils, causing pollution to receiving waters (Aziz et al. 2011).

The technology of solid waste disposal has evolved from conventional to advanced systems which emphasize more on the design, storage capacity and economical principle in receiving various types of wastes including leachate treatment availability. These are the main factors taken into consideration when planning a solid waste disposal site. Above all, proper decisions during designing stage, operation and long-term post-closure plan could ensure efficient monitoring of leachate generation which by far continues to generate even after the landfills have been ceased its operation (Wiszniewski et al. 2006). In general, a landfill will undergo chemical and physical changes caused from the degradation process of solid waste refuse with the soil matrix once the landfilling is complete. Generation of liquid percolates through solid waste matrix assists with rainwater percolation, biochemical, chemical and physical reactions within solid waste refuse directly influencing the quantity and quality of the leachate. In addition, leachate quality and quantity also were influenced by the landfill age, precipitation, weather variation, waste type and composition (Abbas et al. 2009). Principally, a functional landfill site is always occupied with a leachate treatment facility to treat hazardous pollutants in the leachate. Therefore, finding a sustainable method for leachate treatment has always been a priority for landfill managers in order to safely discharge treated leachate into the water bodies without endangering the environment. Over the last decades, new and advanced sustainable technologies of leachate treatment have started received growing interests which offer better removal of leachate pollutants. By utilizing these new technologies, difficult parameters are much easier to treat nowadays. In the early days, landfill leachate was mainly disposed by channeling the leachate pipes to the sewer system and released into the sea. Alternatively, there was also separated system where the leachate pipes were connected with domestic sewage network at conventional sewage plant (Ahn et al. 2002) and treated simultaneously. However, as the volume of leachate

generation increase over time with wide variations in leachate pollutants, this method reduced the treatment efficiency of sewage plant (Çeçen and Aktas 2004). Concerning this, many additional treatments have been proposed and invented in treating landfill leachate separately.

Virtually, various types of treatments have been explored including biological, physical, chemical and physico-chemical techniques. As far as the authors concern, most of the treatments in the market today have their own advantages and limitations. For example, biological treatment is undoubtedly the most effective way in treating high concentration of BOD<sub>5</sub> (Renou et al. 2008). However, depending on the nature of leachate pollutants, sludge bulking may occur in conventional aerobic system which disturbs the leachate treatability (Dollerer and Wilderer 1996). Conventional physico-chemical techniques such as chemical precipitation (Chen et al. 2012; Zhang et al. 2009b; Di Iaconi et al. 2010), adsorption (Ching et al. 2011; Kamaruddin et al. 2011; Lim et al. 2009; Singh et al. 2012), coagulation/flocculation (Liu et al. 2012; Al-Hamadani et al. 2011; Ghafari et al. 2010), chemical oxidation (Sun et al. 2009; Anglada et al. 2011; Cortez et al. 2011a, b) may be used as co-treatment along biological treatments. These techniques have been proven suitable in dealing with difficult parameters in leachate including humic, fulvic acid, heavy metals, adsorbable organically bound halogens (AOXs), polychlorinated biphenyls (PCBs) and several other of persistent organic pollutants (Abbas et al. 2009). Very recently, numerous studies have been introduced which focuses on new and advance treatment. In view of that, various factors have been considered in proposing an ideal treatment system that results in high efficiency of parameters reduction as to comply with the permissible discharge limit enforced by the authorities. Therefore, the purpose of this article aims to summarize leachate sustainable treatment processes including biological, physical and chemical techniques reported from 2008 to 2012. The articles discussed in depth about existing and new treatment methods in treating high concentration of leachate and its progress in the recent years.

### Landfill leachate composition

The leachate generated from the degradation of solid wastes widely varies in terms of composition. Moreover, the risk of obtaining a concentrated leachate depends on a number of factors that control its quantity and quality, such as water percolation through the wastes, biochemical processes in wastes' cell and the degree of wastes compaction (Abbas et al. 2009; Li et al. 2010; Xu et al. 2010). Typically, leachate parameters vary depending on the age of the landfill. For instance, young leachate

(1–2 years) is characterized by high organic fraction of relatively low molecular weight such as volatile organic acids, high COD, total organic carbon (TOC), BOD<sub>5</sub> and a BOD<sub>5</sub>/COD >0.6 (Umar et al. 2010). In contrast, old leachate (>10 years) is characterized by a relatively low chemical oxygen demand (COD) (<4,000 mg/L), slightly basic (pH > 7.5) and low biodegradability (BOD<sub>5</sub>/COD <0.1) (Li et al. 2010). Apart from that, humic and fulvic acid and NH<sub>3</sub>-N as well are greatly produced at this stage due to anaerobic decomposition (Bashir et al. 2011). After landfilling period, BOD<sub>5</sub> content will be degraded during the stabilization stage. Therefore, the BOD<sub>5</sub>/COD ratio decreases with time because the non-biodegradable portion of COD stays unchanged in this process (Ahmed and Lan 2012). Alternatively, climate, landfill cover and type of waste at the landfill site played a major role to the leachate generation rate. A landfill site which is located at hot and arid region tends to generate smaller amount of leachate because of low precipitation whereby, leachate generation is high at tropical weather climate region due to higher precipitation infiltrates into the landfill cell (Renou et al. 2008). Utilization of cover materials during cell development whether as intermediate or final layer is one of the methods in protecting buried refuse on the landfill site to enable biodegradation of solid waste in the refuse. The utilization of impermeable type of cover materials will only increase the confining leachate amount whereby the movement of leachate within the cell is hindered and reduce the effectiveness of landfill cell. In a nut shell, having different leachate characteristics requires in depth understanding of leachate treatability to effectively reduce hazardous pollutants in leachate (Aziz et al. 2011). Table 1 shows typical leachate characteristics from semi-aerobic and anaerobic landfills in Northern Malaysia. Generally, semi-aerobic and anaerobic landfill leachate quality shows wide variation in terms of leachate parameters which indicates that aeration process plays a significant role in lowering several contaminants particularly for the case of Pulau Burung Landfill. Lower ratio of BOD<sub>5</sub>/COD for Pulau Burung Landfill shows that the leachate is in the stabilized stage and difficult to be degraded further biologically (Aziz et al. 2010). In this case, physico-chemical process techniques are mostly recommended for stabilizing leachate (Ghafari et al. 2010). In contrast, the ratio of BOD<sub>5</sub>/COD of 0.205 for Kulim Landfill indicates that the leachate is in the young condition and not in the stabilized stage. Previous works by various researchers (Bashir et al. 2009; Salem et al. 2008; Aghamohammadi et al. 2007) have shown that the ratio of BOD<sub>5</sub>/COD was in the range 0.043 to 0.67 pertaining to various types of landfill leachate that are in agreement with the work by Aziz et al. (2010).

**Table 1** Typical leachate characteristics from Semi-aerobic and anaerobic landfills in Malaysia

Landfill	Semi-aerobic Pulau Burung (aerated)	Anaerobic Kulim (unaerated)	Discharge limit, DOE, Malaysia <sup>a</sup>
Parameters	Average values		–
Phenols (mg/L)	1.2	2.6	–
Ammonia-N	483	300	–
Total nitrogen (mg/L)	542	538	–
Nitrate-N (mg/L)	2,200	1,283	–
Nitrite-N (mg/L)	91	52	–
Total phosphorus (mg/L)	21	19	–
Orthophosphate (mg/L)	141	94	–
BOD <sub>5</sub> (mg/L)	83	326	50
COD	935	1,892	100
BOD <sub>5</sub> /COD	0.09	0.205	0.5
pH	8.2	7.76	5.5–9
Turbidity (NTU)	1,546	8.55	–
Color	3,334	1,936	–
Total solids (mg/L)	6,271	4,041	–
Suspended solids (mg/L)	1,437	6,336	–
Total iron (mg/L)	7.9	707	100
Zinc (mg/L)	0.6	5.3	5
Total coliform	–	0.2	1
<i>E. Coli</i>	–	$0.81 \times 10^{-4}$	–

Adapted from Aziz et al. (2010)

<sup>a</sup> Second schedule (Regulation 13), amended 2013: Acceptable conditions for discharge of leachate

### Leachate treatment techniques

Satisfactorily knowledge in landfill leachate characteristics is required to understand the variable performance found in treating the leachate either by biological, physical or physico-chemical methods. In the last few years, biological treatment has attracted more interests due to its many advantages which includes variety of sources and the ease and speed which the microorganisms can be cultured and produced (Zhao et al. 2010). These systems are divided into aerobic (with oxygen) and anaerobic (without oxygen) conditions. In particular, the use of microorganisms or bacteria to remove the contaminants in leachate is through assimilating process. This process helps to increase microbial metabolism and building blocks of the living cell. As a result, the metabolic conditions of the living cells

are capable to remove leachate parameters. Regardless of the choice of application, an appropriate selection of biological treatment requires ample thought for cultivating and maintaining an acclimated healthy biomass, flow rate tolerance and organic loads to be treated. Until now, biological treatments are still one of the acceptable means in treating leachate because it offers low capital and operating cost to the operators. In addition, the application of biological treatment has been proven a total destruction of organic, sulfides, organic compounds, and toxicity.

Biological treatment has been shown very effective in removing organic and nitrogenous matter (Abbas et al. 2009) including immature leachate when the BOD<sub>5</sub> concentration is high and the BOD<sub>5</sub>/COD ratio is more than 0.5 (Renou et al. 2008). However, as the biodegradation of solid waste progress, the efficiency of biological process reduces due to the increasing amount of refractory compounds namely fulvic and humic acids constituents in leachate. Nevertheless, simplicity, ease of operation and reliability have been the methods of choice in employing biological process in the early days of landfill leachate treatment process (Renou et al. 2008). In this section, we summarized a few suspended and attached growth systems that are commonly used in leachate treatment such as batch reactor, bioreactor, growth plant and microbial consortium, and combination of biological devices. These techniques, although have been seen as conventional practices, are still reliable in treating high BOD<sub>5</sub> contents in the landfill leachate particularly for landfill categorized as young and intermediate class. Table 2 shows some of the selection of biological treatment, their criteria and application method in a simplified format.

## Biological process

### Batch reactor

Xu et al. (2010) performed a partial nitrification, aerobic ammonium oxidation (Anammox) and heterothopic denitrification by sequencing batch reactor (SBR). The experimental conditions of  $30 \pm 1$  °C and dissolved oxygen (DO) of concentration within 1.0–1.5 mg/L were fixed in the SBR. They found that maximum aerobic ammonium oxidizing and anaerobic ammonium oxidizing are achieved at 0.79 and 0.18 (kg –N/kg<sub>dw</sub>/day) after the inoculation of Anammox biomass and aerobic activated sludge (80 % w/w) that last for 86 days. In contrast, aerobic ammonium oxidizing, anaerobic ammonium oxidizing and denitrification reached 2.83, 0.65 and 0.11 (kg –N/kg<sub>dw</sub>/day) when denitrifying bacteria was inoculated into the reactor along with the feeding of raw landfill leachate. In other study, Spagni and Marsili-Libelli (2009) focused on the nitrification and denitrification processes of stabilized leachate by SBR

process to enhance the nitrogen removal efficiency. They reported that by adding external COD and adjusting the length of oxic phase could increase nitrogen rate removal. Meanwhile, Lan et al. (2011) successfully conducted simultaneous partial nitrification anammox and denitrification (SNAD) process by SBR which focused on the influence of hydraulic retention time (HRT). They concluded that increasing the HRT from day 3 to 9 of SBR process would increase the COD (87–96 %). Meanwhile, different observations were recorded when pH and DO were reduced which result in lower removal of COD and nitrogen. Finally, they revealed total nitrogen (TN) removal of 85–87 % by anammox with partial nitrification and 7–9 % by denitrification from the SNAD process, respectively. Aziz et al. (2011) utilized SBR instruments for the swim-bed biofringe process for the removal of COD, BOD<sub>5</sub>, TKN and NH<sub>3</sub>-N from stabilized leachate. They utilized activated sludge and biofringe as the main process parameters. The results demonstrated that swim-bed BF was capable of removing nitrite, nitrate and phosphorus from leachate. On the contrary, the removal performance for COD and NH<sub>3</sub>-N was not significant, respectively.

### Bioreactor

Yahmed et al. (2009) conducted an investigation of a pilot unit system consisting of three unit fixed bioreactors. They tested for different organic loading rate (OLR) of microbials namely *Actinomyces*, *Bacillus*, *Pseudomonas* and *Burkholderia* for the removal of TOC. They concluded that the maximum TOC reduction by *Pseudomonas* isolates was of 70 %. Meanwhile, *Actinomyces* isolates, *Bacillus* isolates and *Burkholderia* isolates gave 69, 69 and 77 % TOC reduction, respectively. In another study, Ellouze et al. (2008) investigated leachate treatability by utilizing sludge from a waste water treatment plant. Preliminary studies showed that the acclimatization of the sludge was able to remove organic matter and toxicity. A set up of stirred tank reactor with OLR from 0.5 to 4 g/L/day with HRT decreased from 50 to 4.6 days demonstrating that COD was removed up to 80 % for a loading rate of 5.4 g/L/day. In addition, the concentration of N-NH<sub>4</sub><sup>+</sup> was reduced below to the recommended standard. Finally, the results from toxicity of *Vibrio fischeri* and the germination of *Lepidium sativum* seeds showed that the treatment was able to effectively provide detoxification of the effluent whereby the loading rate up to 6 g/L was ideal for the perturbation of the system which triggered an accumulation of residual COD and toxicity, respectively. Ismail et al. (2011) investigated the effect of different organic loading charges (0.6–16.3 kg) for the removal of TOC and TKN by submerged biofilm reactor. The results showed that without

**Table 2** Biological treatments and method of application

Biological treatment	Common experimental condition	Example of work		
		Experimental handling	Parameters concern	Reference
Anaerobic filter/digester/reactor	<ul style="list-style-type: none"> <li>Emit biogas (CH<sub>4</sub>, CO<sub>2</sub>)</li> <li>Tolerable to high COD</li> <li>Good precipitation for toxic metals</li> </ul>	Used seed sludge as inoculate	COD, pH, Al, Fe, Zn, Ni, Cd, Mn, Pb, Cu and Cr NH <sup>4+</sup>	Kawai et al. (2012b)
		Used activated sludge as end treatment	COD, BOD <sub>5</sub> and TSS	Kheradmand et al. (2010)
		Co digester of leachate and sewage sludge	Biomethanation production (BMP) volatile solids reduction (VSR)	Hombach et al. (2003)
		Anaerobic sludge used as inoculums	COD, CH <sub>4</sub>	Imen et al. (2009)
Upflow anaerobic sludge blanket (UASB)	<ul style="list-style-type: none"> <li>Normal UASB works with anaerobic bacteria</li> </ul>	Mature leachate was co-digested with synthetic waste water	COD, CH <sub>4</sub>	Kawai et al. (2012a)
Aerated lagoons	<ul style="list-style-type: none"> <li>Aerobic condition on top of lagoon</li> <li>Anaerobic condition at the lower</li> <li>High and low speed aerators used to disperse water into droplet to allow oxygen enter</li> </ul>	Four connected aerated lagoons	COD, NH <sup>4+</sup>	Mehmood et al. (2009)
Activated sludge plants/reactor	<ul style="list-style-type: none"> <li>Sludge contents is higher than aerated lagoon, possible for short residence time</li> </ul>	Pre-denitrification activated sludge with bentonite additive	COD, NH <sub>3</sub> -N	Wiszniowski et al. (2006)
		Phase separation through aeration	COD, BOD <sub>5</sub> , NH <sup>4+</sup> and total nitrogen	Jun et al. (2007)
Rotating biological contactors (RBC)	<ul style="list-style-type: none"> <li>Bacteria attached to the contactors</li> <li>Suitable for low organic content in leachate</li> </ul>	Utilized single-stage anoxic RBC	NO <sup>3-</sup>	Cortez et al. (2011a, b)
Biological co-treatment	<ul style="list-style-type: none"> <li>Combination of reactor</li> <li>Denitrifying reactor</li> <li>Reactors with denitrifying and methanogenesis</li> <li>Partial nitrification, anaerobic ammonium oxidation (anammox) and heterotrophic denitrification</li> <li>Aerobic and anaerobic condition in a reactor</li> <li>Selection of disc for cyclic bath RBC</li> <li>Different hydraulic retention times (HRT), rotational speeds, and with varying organic concentrations</li> </ul>	Simultaneous aerobic and anaerobic (SAA) bioreactor	COD	Yang and Zhou (2008)
		Landfill simulate reactor plus activated sludge reactor	COD, NH <sup>4+</sup>	Shou-liang et al. (2008)
		Two stage UASB and anoxic-oxic reactor	COD, BOD <sub>5</sub>	Peng et al. (2008)
		Aerobic activated sludge as inoculums and SBR as the experimental reactor	NH <sup>4+</sup>	Xu et al. (2010)
		Leachate recirculate plus anaerobic and aerobic of msw	pH, alkalinity, total dissolved solids, conductivity, oxidation-reduction potential, chloride, chemical oxygen demand, ammonia, and total Kjeldahl nitrogen, in addition to generated leachate quantity	Bilgili et al. (2007)
		RBC and upward-flow anaerobic sludge bed reactor	COD	Castillo et al. (2007)

initial pH adjustment, TOC removal rate varied between 65 and 97 %. The total reduction of COD reached 92 % at a HRT of 36 h. However, the removal of total Kjeldahl nitrogen for loading charges of 0.5 kg N/m<sup>3</sup>/day reached 75 %. Further toxicity test for the removal of organic carbon and nitrogen showed that *Bacillus*, *Actinomyces*, *Pseudomonas* and *Burkholderia* genera were responsible for these occurrences. Chen et al. (2008) investigated the performance of a moving bed biofilm reactor (MMBR) via aerobic and anaerobic sequence for simultaneous removal of COD and ammonium. They discovered that anaerobic MBBR played a major role in COD removal (91 %) at OLR of 4.08 kg COD/m<sup>3</sup>/day due to methanogenesis and the aerobic MBBR acted as COD-polishing and ammonium removal step. In contrast, HRT at 1.25 days required to remove more than 97 % of NH<sub>4</sub><sup>+</sup> of the aerobic MBBR. Bohdziewicz et al. (2008) examined the treatability of leachate by submerged membrane bioreactors. They used synthetic waste water as feeding medium by volume ratio with the addition of leachate dilution between 50 and 75 %. They claimed that higher COD removal could be achieved with the leachate addition of 10–20 % v/v. They also revealed that the best anaerobic digestion efficiency (COD removal 90 %) was observed for HRT for 2 days and OLR of 2.5 kg COD/m<sup>3</sup> days for the optimal anaerobic digestion efficiency.

#### Growth plant and microbial consortium

Ye et al. (2008) tested immobilized microbial for the removal of COD and NH<sub>3</sub>-N. They measured the efficient microbial flora on the carrier by Kjeldahl's method. The biological process showed that immobilized microorganisms system was effective for the removal of COD and nitrogen at 98.3 and 99.9 %, respectively. A study done by Saetang and Babel (2012) revealed that *Trametes versicolor* BCC 8725 could remove 78 color, 68 BOD<sub>5</sub> and 57 % COD from leachate sample within 15 days at optimum condition, respectively. They also claimed that organic loading and ammonia were the factors that affected the biodegradation. In another work, Białowiec et al. (2012) compared reed and willow with an unplanted control by measuring redox potential levels in the *rhizosphere* of microcosm system for the leachate bioremediation. The results suggested that redox potential in the reed *rhizosphere* was anoxic (mean  $-102 \pm 85$  mV), but it was the least negative, being significantly higher than in the willow (mean  $-286 \pm 118$  mV), which had the lowest Eh. They also reported that NH<sub>4</sub><sup>+</sup> reduced from the first day and remained at a similar low level until 4 weeks of the experimental period. Meanwhile, Loncnar et al. (2010) discovered that the planted willows at a recirculation

process of leachate showed a high sustainability of saline ions. The concentration of saline ions was recorded at ranges 132 to 2,592 mg Cl<sup>-</sup>/L, 69 to 1,310 mg Na<sup>+</sup>/L and 66 to 2,156 mg K<sup>+</sup>/L, with mean values of 1,010, 632 and 686 mg/L, respectively. Akinbile et al. (2012) found that by utilization of *Cyperus haspan* with sand and gravel in a constructed wetland with optimum retention time of 3 weeks could efficiently reduce heavy metals parameters at the ranges of 33–89 %. Meanwhile, significant reduction of TSS, COD, BOD<sub>5</sub>, NH<sub>3</sub>-N, and TP of 98, 92, 79, 54 and 99 % was recorded, respectively. In another work, using anaerobic organisms in a series of anaerobic tanks filled with leachate, 100 mL of anaerobic organism and 14 days of microbial inhibitors, 65.5, 60.2 and 46.3 % of COD, NH<sub>3</sub>-N and color were removed, respectively (Kamaruddin et al. 2013).

#### Physical–chemical process

Generally, satisfactory treatment of landfill leachate is dependent on methods applied to leachate generation handling. A complete landfill leachate treatment usually consists of physical, chemical and biological processes. Physical treatment utilizes non chemical or biological changes in the leachate whereby only physical phenomenon is used to enhance leachate quality. For example, screening of leachate is done by employing metal grit trap to retain larger impurities prior to subsequent treatment. Meanwhile, sedimentation process is involving settling of solids by gravitational force by simply allowing short residence time in sedimentation tank. This process is crucial for flocs formation. Another type of physical treatment is aeration which utilizes oxygen as the oxidation agent in leachate lagoon. This process has been found to enhance the removal of BOD<sub>5</sub> in pre-treatment as proven by many successful treatment selections. In contrast, chemical treatment utilizes chemicals additive that involves reaction to improve leachate quality. During chemical treatment, neutralization is commonly used to neutral leachate condition by the addition of acid or base in the process. In other process, coagulation has been known as one of the oldest chemical treatment in landfill leachate treatment. It utilizes chemical additives which enable the formation of insoluble end products and capable of removing a wide range of leachate parameters through ionic mechanism. In addition, certain types of polyvalent metals are widely used as coagulant or coagulant aid such as ferric chloride, polyaluminum chloride, aluminum sulfate or ferric sulfate. Alternatively, disinfection of leachate is one of established methods in chemical treatment. Chlorine known as the strong oxidizing agent is commonly used to kill bacteria when crucial biological process is affected by the chlorine. In a nut shell, physical–chemical process, includes

adsorption, coagulation/flocculation and chemical oxidation, is commonly used when the biological process is hindered due to excessive presence of refractory compounds in leachate. Normally, physical–chemical process is carried out as a pre-treatment or at the final stage of the leachate treatment process. Table 3 discusses the criteria of the most common biological and physical–chemical process in leachate treatment and their advantages.

#### Activated carbon adsorption

Adsorption of leachate by activated carbon has received great interests considering its superior properties having larger surface area, high adsorption capacity and better thermal stability. Ching et al. (2011) used a chemically treated coffee ground-activated carbon for the removal of total iron and orthophosphate from stabilized leachate. They discovered that optimum removal for the latter was attained at impregnation ratios (IRs) of 2.5 and 0.5 at doses of 10 g and pH 8.1. In contrast, pH 13 was found optimum for total iron removal while pH < 5 and >11 was optimum for PO<sub>4</sub>-P removal. Kamaruddin et al. (2011) concluded that the optimum preparation conditions of durian peel-activated carbon (DPAC) was achieved at IR, activation temperature, and activation time of 3, 400 °C and 2.2 h, for the removal of NH<sub>3</sub>-N from stabilized leachate. The optimum conditions of DPAC are capable of removing 47 % of NH<sub>3</sub>-N. Kalderis et al. (2008) investigated ZnCl<sub>2</sub>-treated rice husk and sugarcane bagasse-activated carbons. The activated carbons were tested for humic acid, phenol and leachate parameters removal. They found that both ACs showed the best

adsorption behavior towards phenol, removing around 80 % at 4 h equilibrium period. However, the adsorption for arsenic and humic acids was lower than that of phenol based on isotherm data. Finally, they revealed that with 30 g/L of AC, it was possible to remove 70 and 60 % of COD and color, respectively. Singh et al. (2012) developed isotherm and kinetic models for three types of commercially available activated carbons. They suggested that Redlich–Peterson model showed better fit to the experimental data and the TOC adsorption capacity for both micro-porous and meso-porous activated carbons. In addition, intraparticle diffusion coefficients (De) for both AC were in the order 10<sup>-10</sup> m<sup>2</sup>/s for particle sizes >0.5 mm. Lim et al. (2009) established an axial dispersion model for palm shell-activated carbon (PSAC) in column mode. The applicability of the model was tested for the removal of COD and turbidity of leachate. The highest breakthrough of COD was obtained at Empty Bed Contact Time (EBCT) of 14.7 min, with sorption capacity of 1,460 mg/g. In contrast, turbidity and pH effluent showed insignificant effect on EBCT, respectively.

While activated carbon has gained much popularity in the market nowadays, there is also several type of adsorbents receiving great interest in the recent years due to their abundance, easily obtained, high regeneration cycle, and higher mechanical stability in adsorption studies. Accordingly, waste materials such as from agricultural sectors and industrial byproducts have been identified to have the potential as an alternative adsorbent in adsorption studies. Table 4 shows several types of adsorbents that have been proposed and tested in treating landfill leachate by adsorption studies.

**Table 3** Criteria of biological and physical–chemical treatment

Treatment option	Treatment process	Treatment efficiency			Operational cost	Space requirement
		Leachate condition				
		Young leachate	Medium age leachate	Mature leachate		
Biological	Rotating biological contactor (RBC)	Strong	Fair	Weak	Expensive	Normal
	Sequencing batch reactor (SBR)	Strong	Fair	Weak	Moderate	Normal
	Moving bed biofilm reactor (MBBR)	Strong	Fair	Weak	Expensive	Large
	Membrane bioreactor (MBR)	Strong	Fair	Weak	Expensive	Large
	Upflow anaerobic sludge blanket (UASB)	Strong	Fair	Fair	Moderate	Normal
Physical-chemical	Activated sludge	Strong	Fair	Weak	Expensive	Large
	Phytoremediation	Fair	Fair	Good	Inexpensive	Large
	Lagooning	Strong	Fair	Weak	Expensive	Large
	Adsorption	Weak	Fair	Weak	Expensive	Normal
	Coagulation	Weak	Fair	Fair	Inexpensive	Medium
	Chemical oxidation	Weak	Fair	Fair	Expensive	Normal
	Stripping	Weak	Fair	Fair	Expensive	Large
	Precipitation	Weak	Fair	Fair	Inexpensive	Medium

**Table 4** New type of adsorbents used in landfill leachate adsorption

Adsorbent	Source	Parameters concern	Reference
Turkish clinoptilolite	Local supply	Ammonium	Karadag et al. (2008)
Ion resins	Local supply	Color, COD, NH <sub>3</sub> -N,	Bashir et al. (2010)
Kemiron	Local supply	Arsenic	Oti et al. (2011)
Honeycomb cinders	Byproducts from briquette combustion	PO <sub>4</sub> -P, COD	Yue et al. (2011)
Sphagnum peat moss	Local supply	Cd, Ni	Champagne and Li (2009)
Crushed mollusk shells	Local supply	Cd, Ni	
Composite adsorbent	Local supply and Agri-wastes	NH <sub>3</sub> -N, COD	Halim et al. (2010b)
Limestone, granular AC	Local supply	Orthophosphate	Hussain et al. (2011)
Activated carbon, bone meal and iron fines	Local supply	Al, As, Ca, Cd, Co, Cr, Cu, Fe, Hg, Mg, Mn, Mo, Ni, Pb, Sr and Zn	Modin et al. (2011)
Coal fly ash	Thermal power plant	Zn, Pb, Cd, Mn and Cu	Mohan and Gandhimathi (2009)
Durio zibethinus L.	Agricultural waste	NH <sub>3</sub> -N, carbon yield	Kamaruddin et al. (2011)

### Coagulation flocculation

Coagulation and flocculation is known as one of the oldest treatment methods in landfill leachate. Apart from that, it has been widely used in treating stabilized (Al-Hamadani et al. 2011) and matured landfill leachate (Vedrenne et al. 2012). In addition, the application of coagulation and flocculation can be used as pre-treatment process in order to remove non-biodegradable organic matter (Renou et al. 2008). Several studies have identified the selection of appropriate experimental conditions when employing coagulant and flocculation process. Ghafari et al. (2009) used PAC and alum to treat stabilized leachate in coagulation/flocculation process at maintained mixing time and mixing speed. They utilized CCD and RSM to establish the relationship between operating variables (dosage and pH) and leachate parameters removal. The results indicated that the optimum conditions for PAC was obtained at dosage of 2 g/L and pH 7.5 which managed to reduce COD, turbidity, color and TSS concentrations at 43.1, 94.0, 90.7, and 92.2 %. Subsequently, the optimum condition for alum was achieved at dosage 9.5 g/L and pH 7 which further reduced COD, turbidity, color and TSS concentrations to 62.8, 88.4, 86.4, and 90.1 % respectively. However, when they optimized the speed and time for rapid and slow mixing, they observed that COD removal was achieved at 84.5 and 56.7 % for alum and PAC. Single use of PAC showed that 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 %) (Ghafari et al. 2010). Liu et al. (2012) used RSM for the optimization process of polyferricsulphate (PFS) coagulant towards COD, color, turbidity and HA removal. At optimum conditions, COD, color, turbidity and HA removal of 56.38, 63.38, 89.79, 70.41 % were observed at PFS dose of 8 g/L at pH 6.0, FeCl<sub>3</sub>·6H<sub>2</sub>O dose of 10 g/L at pH 8.0 and

Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>·7H<sub>2</sub>O dose of 12 g/L at pH 7.5. Using similar optimum variable conditions, 68.65, 93.31, 98.85, 80.18 % for FeCl<sub>3</sub>·6H<sub>2</sub>O and 55.87, 74.65, 94.13, 53.64 % for Fe<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>·7H<sub>2</sub>O of COD<sub>cr</sub>, color, turbidity and HA removal were observed, respectively. In another study, an alternative coagulant was successfully synthesized and tested. Al-Hamadani et al. (2011) compared psyllium husk as coagulant aid with PACl and alum. They found that the maximum removal was achieved when psyllium husk was used as coagulant aid with PACl resulting in COD, color and TSS removal of 64, 90 and 96 %, respectively. Meanwhile, Syafalni et al. (2012) compared lateritic soil coagulant with alum in jar test experiment. The optimum condition was achieved at pH 2 and lateritic soil coagulant dose of 14 g/L resulting 65.7 % COD, 81.8 % color and 41.2 % NH<sub>3</sub>-N removal. Comparable finding was observed when alum was used at pH 4.8 and coagulant dosage of 10 g/L where COD, color and NH<sub>3</sub>-N were removed at 85.4, 96.4 and 47.6 %, respectively. Tzoupanos et al. (2008) evaluated the performance of polyaluminium silicate chloride (PSC) coagulant with different Al to Si molar ratio with biologically treated leachate. The results suggested that PSC had better removal of COD and color than PACl due to high tolerance against pH ranging from 7 to 9. Concerning with the inhibitory of dissolved organic matters, Comstock et al. (2010) compared three types of coagulants which focused on dissolved organic matter (DOM) removal from leachate. The presence of DOM was measured using specific ultraviolet (UV) absorbance at 254 nm (SUVA<sub>254</sub>) and fluorescence excitation–emission matrices. The performance of the metals salts was in the order of: ferric sulfate > aluminum sulfate > ferric chloride and DOM removal followed the trend of color > UV<sub>254</sub> > dissolved organic carbon > COD. In another study, Yimin et al. (2008) used poly-magnesium–aluminum sulfate (PMAS) in jar test experiment. The



removal of COD, BOD<sub>5</sub>, UV<sub>254</sub>,(OM) by PMAS was observed at 65, 60, 85 % under optimum conditions, respectively.

#### Chemical oxidation

Generally, chemical oxidation process utilizes chemical substances, mainly chlorine, ozone, potassium permanganate and calcium hydroxide (Abbas et al. 2009). In addition, advance oxidation process (AOP) normally is used to enhance the chemical oxidation efficiency to the stable oxidation state. Owing to the successful rate of the removal of refractory compounds in leachate, AOP, however, has some limitation including high energy requirement, and chemical reagent (Kalderis et al. 2008) throughout the leachate treatment process. Nevertheless, AOP still considered as the better treatment methods when employing it as pre-treatment prior to the biological process thereby reducing capital operation of leachate treatment. Previous studies have demonstrated that chemical precipitation, Fenton/Electro-Fenton/Photo-Fenton, Photochemical/Photoelectrochemical/Photocatalytic could significantly reduce leachate containing refractory compounds. These processes include both non photochemical and photochemicals which generate hydroxyl radicals with and without light energy (Wiszniewski et al. 2006). Table 5 summarizes some of the major breakthroughs in the utilization of AOP techniques which results in significant removal of leachate pollutants.

#### Advanced biological/physical–chemical techniques

With stringent requirement by authorities in protecting environmental fate, the treatability of landfill leachate is a prominent challenge for the landfill operator to comply with the current regulations. With regards to this, conventional treatment is not sufficient to render high concentration of leachate pollutants. Therefore, the adverse impacts of inefficient leachate treatment have raised serious concerns to the society and environment, respectively. Ultimately, the combination of individual treatment process into hybrid process has been more effective and emerged as the choice of treatment for landfill operators. Kwon et al. (2008) found that higher reduction of COD<sub>cr</sub>, color and TP could be achieved when they employed nanofiltration-rotary disk membrane (NF-RDM) process. In addition, the introduction of RO with NF-RDM process enhanced NH<sub>4</sub><sup>+</sup> removal from 25 to 92 %. In another study, Tsilogeorgis et al. (2008) concluded that ultrafiltration membrane-SBR was able to remove TN removal (88 % maximum) over 4 months monitoring. However, COD removal varied (40–60 %) due to high SRT. Also, PO<sub>4</sub>-P removal efficiency was varied (35–45 %) during the

first 50 days of operation due to direct addition of KH<sub>2</sub>PO<sub>4</sub>/K<sub>2</sub>HPO<sub>4</sub> that was aimed to improve C:N:P ratio.

In a hybrid experimental work, Li et al. (2010) investigated coagulation/flocculation augmented powdered activated carbon (PAC). They used four types of commercially available coagulants to determine optimum working conditions and found that PFS showed better removal for COD, SS, turbidity, toxicity and sludge volume at 70, 93, 97 % and 32 mL. Consequently, 10 g/L of PAC was found optimum with 90 min contact time during experimental period. Under optimum conditions of combined techniques, COD, Pb, Fe and toxicity removals were found 86, 97.6, 99.7 and 78 %, respectively. Meanwhile, to improve pollutants removal, Palaniandy et al. (2010) found that the combination between FeCl<sub>3</sub> coagulation and dissolved air flotation (DAF) managed to reduce turbidity, COD, color and NH<sub>3</sub>-N concentration up to 50, 75, 93 and 41 %. The statistical analysis suggested that the optimum operating conditions for coagulation and DAF were 599.22 mg/L of FeCl<sub>3</sub> at pH 4.76 followed by saturator pressure of 600 kPa, flow rate of 6 L/min and injection time of 101 min. In another work, Poznyak et al. (2008) injected ozone process after the coagulation/flocculation treatment. They found that coagulation/flocculation injected ozone could remove 70 % of humic substances in leachate. Next, when ozone process was further induced, color was 100 % removed during 5 min period. Finally, they found that organic substance diminished completely during 15 min ozonation when extracted with chloroform-methanol and 5 min when extracted with benzene. Ying et al. (2012a) applied various treatment processes with combination of internal micro-electrolysis (IME) without aeration and IME with full aeration in one reactor. The authors implemented a novel sequencing batch internal micro-electrolysis reactor (SIME) throughout the experimental work. Results showed that high COD removal efficiency of 73.7 ± 1.3 % was obtained which was 15.2 and 24.8 % higher than that of the IME with and without aeration, respectively. The SIME reactor also exhibited a COD removal efficiency of 86.1 ± 3.8 % to mature landfill leachate in the continuous operation, which was much higher ( $p < 0.05$ ) than that of conventional treatments of electrolysis (22.8–47.0 %), coagulation–sedimentation (18.5–22.2 %), and the Fenton process (19.9–40.2 %), respectively (Ying et al. 2012b).

Among advanced oxidation processes, several improvements towards the capabilities of existing techniques have been explored by various authors. Galeano et al. (2011) utilized catalytic wet peroxide oxidation (CWPO) with an Al/Fe-pillared clay catalyst in semi-batch reactor. The COD was found reduced up to 50 % and biodegradability index (BI) output was exceeding 0.3 during 4-h experiment duration. They concluded that high

**Table 5** Summary of advanced oxidation process focused with mediator and its parameters

Process	Mediator	Parameter concerns	Removal (%)	Reference
<b>Chemical precipitation</b>				
	Magnesium ammonium phosphate (MAP),	NH <sup>4+</sup>	95	Xiu-Fen et al. (2011)
		COD	56	
	MAP	NH <sup>4+</sup>		Zhang et al. (2009b)
	MAP	NH <sup>4+</sup>	>95	Di Iaconi et al. (2010)
	Magnesit (MgCO <sub>3</sub> ) during MAP precipitation	NH <sup>4+</sup> PO <sub>4</sub> <sup>3-</sup> Turbidity		Gunay et al. (2008)
	Sodium hypochlorite	<i>Escherichia coli</i>	99	Umar et al. (2011)
<b>Fenton/Electro-Fenton/Photo-Fenton</b>				
Fenton and photo-Fenton		COD TOC	70	Hermosilla et al. (2009)
Electro-Fenton	Cathode was nourished with oxygen that submitted to H <sub>2</sub> O <sub>2</sub> electrochemically	TOC Total nitrogen (TN)	82 51	Wang et al. (2012)
Electro-Fenton		COD Color PO <sub>4</sub> <sup>3-</sup> NH <sub>4</sub> -N	72 90 87 28	Atmaca (2009)
Fenton	Fe <sup>2+</sup> and Fe <sup>3+</sup> as catalyst and H <sub>2</sub> O <sub>2</sub> as oxidizing agent	COD Color	58.3 78.3	Mohajeri et al. (2010)
Fenton	FeCl <sub>2</sub>	BOD <sub>5</sub> /COD	(0.05–0.21)	Zhang et al. (2009a)
<b>Photochemical/photoelectrochemical/photocatalytic</b>				
Photocatalysis	TiO <sub>2</sub>	COD Lead NH <sub>3</sub> -N	86 91 90	Meeroff et al. (2012)
Photoelectrochemical	DSA anode and UV	COD TOC NH <sup>4+</sup>	74.1 41.6 94.5	Zhao et al. (2010)
UV-TiO <sub>2</sub> photocatalysis		COD DOC Color BOD <sub>5</sub> /COD	60 70 97 (0.09–0.39)	Jia et al. (2011)
Photocatalytic	Heterogeneous (TiO <sub>2</sub> /UV, TiO <sub>2</sub> /H <sub>2</sub> O <sub>2</sub> /UV) Homogenous (H <sub>2</sub> O <sub>2</sub> /UV, Fe <sup>2+</sup> /H <sub>2</sub> O <sub>2</sub> /UV)	DOC Aromatic contents		Rocha et al. (2011)
<b>Electrochemical/electro-oxidation/electrocoagulation</b>				
Electrochemical oxidation	Ti/IrO <sub>2</sub> -RuO <sub>2</sub> anode in the presence of HClO <sub>4</sub>	COD TC TP Color	75 90 65 100	Turro et al. (2011)
Electro-oxidation	Boron-doped diamond anode	Color COD NH <sub>3</sub> -N	84 51 32	Anglada et al. (2011)

**Table 5** continued

Process	Mediator	Parameter concerns	Removal (%)	Reference
Electro-oxidation	RuO <sub>2</sub> and IrO <sub>2</sub> as the anode	COD NH <sub>3</sub> -N	33.6–66.4 11.9–98.4	Zhang et al. (2011)

catalyst, low peroxide concentrations, dosages and addition rates were the main factors affecting oxidizing agents in terms of BI and COD removal efficiency. Xu et al. (2012) found that by applying catalytic wet air oxidation (CWAO) with the presence of AC as catalyst and potassium persulfate (K<sub>2</sub>S<sub>2</sub>O<sub>8</sub>) as promoter, almost complete fulvic acid (FA) and COD removal up to 78 % could be achieved in the K<sub>2</sub>S<sub>2</sub>O<sub>8</sub>/AC system at 150 °C and 0.5 MPa oxygen pressure. They also found that the BOD<sub>5</sub>/COD ratio increased from 0.13 to 0.95 after CWAO. Sun et al. (2009) compared the application of Fenton and Oxone/Co<sup>2+</sup> oxidation processes. When they tested Fenton oxidation as standalone process, COD removal was found at 56.9 % but SS and color increased in concentration due to high generation of ferric hydroxide sludge. Subsequently, when they assessed the performance of Oxone/Co<sup>2+</sup> oxidation, the removal of COD, SS and color removal increased to 57.5, 53.3 and 83.3 %. The optimum conditions of the process were: [Oxone] = 4.5 mmol/L, [Oxone]/[Co<sup>2+</sup>] = 104, pH = 6.5, reaction temperature = 30 ± 1 °C, reaction time = 300 min, number of stepwise addition = 7. Panizza et al. (2010) utilized anodic oxidation using electrolyte flow cell equipped with lead dioxide (PbO<sub>2</sub>) anode and stainless steel as cathode. They observed that the galvanostatic electrolyses enhanced COD removal along with rising current, solution pH and temperature. Gabarró et al. (2012) studied the effects of temperature on NH<sub>3</sub>-N in a partial nitrification (PN)-SBR. The stable PN was achieved with minimum volume of 111 L and N-NH<sub>4</sub><sup>+</sup> of 6,000 mg/L at 25 and 35 °C. The result was demonstrated by kinetic model where NH<sub>4</sub><sup>+</sup> and NO<sub>2</sub> concentrations were similar at both temperatures. In contrast, free ammonia and free nitrous acid (FNA) were found differed due to the strong temperature dependence. There are concerns with excessive pollutants concentration in matured leachate,

## Conclusions

Over the years, various sustainable landfill leachate treatment techniques have been proposed and tested for treating highly polluted leachate. At this point, here are some of the key points from the extensive discussions regarding sustainable landfill leachate treatment:

- Refractory compounds in leachate always change over times due to overwhelmed mankind activities. Therefore, modification of existing treatment technique may be viable to ensure that the treatment efficiency is consistent and in accordance to the regulatory standards;
- there has been a steady progress of new and advanced sustainable landfill leachate treatment which proven to be a promising alternative;
- utilization of advanced waste disposal method such as incineration and recycling may be suitable to mitigate the generation of landfill leachate.
- Though there are still uncertainties whether these techniques could enhance environmental sustainability and safety of human being, more efforts should be carried out to ensure a livelihood of human being and earth coexistence;
- therefore, a holistic approach is essential for finding a suitable leachate treatment opportunity in order to safeguard environmental and human being livelihood, as a whole.

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