water

Review

Physicochemical Technique in Municipal Solid Waste (MSW) Landfill Leachate Remediation: A Review

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Abstract: Leachate generation is among the main challenging issues that landfill operators must handle. Leachate is created when decomposed materials and rainwater pass through the waste. Leachate carries many harmful pollutants, with high concentrations of BOD, COD, colour, heavy metals, ammoniacal nitrogen (NH_3-N), and other organic and inorganic pollutants. Among them, COD, colour, and NH_3-N are difficult to be completely eliminated, especially with a single treatment. They should be handled by appropriate treatment facilities before being safely released into the environment. Leachate remediation varies based on its properties, the costs of operation and capital expenditures, as well as the rules and regulations. Up until now, much scientific and engineering attention was given to the development of comprehensive solutions to leachate-related issues. The solutions normally demand a multi-stage treatment, commonly in the form of biological, chemical, and physical sequences. This review paper discussed the use of contemporary techniques to remediate landfill leachate with an emphasis on concentrated COD, colour, and NH_3-N levels with low biodegradability that is normally present in old landfill or dumping grounds in developing countries. A semi-aerobic type of landfill design was also discussed, as this concept is potentially sustainable compared to others. Some of the challenges and future prospects were also recommended, especially for the case of Malaysia. This may represent landfills or dumpsites in other developing countries with the same characteristics.

Keywords: physicochemical; solid waste; COD; colour; landfill; leachate; semi-aerobic

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1. Introduction

The number of residents in Malaysia in the third quarter of 2022 was reported to be 32.9 million [1]. This results in a massive quantity of municipal solid waste (MSW) production of around 38,427 metric tonnes per day (1.17 kg/capita/day). A total of 82.5% of waste ends up in landfills. In the same year, the annual MSW collection reached 14 million metric tonnes [2]. Organic waste stands between 40% and 60% of waste in most developing countries. Although there are many advantages to landfilling for waste disposal, it raised serious issues because of the highly polluted leachate it creates. Leachate contaminants include more than 200 different chemicals, most of which are hazardous to the environment. Rainwater infiltrating the deposited waste at the landfill or dumpsite results in the formation of leachate. Additionally, leachate may be produced from a number of different sources, such as transpiration, groundwater intake, storing wet materials, evaporation losses, surface flow, and the hydrolysis and biodegradation of organic molecules [3]. Leachate may drain as runoff or move to the bottom of the waste body. These may pollute groundwater and surface water, endangering both aquatic life and human health. The amount and composition of leachate are influenced by a variety of variables, including seasonal weather fluctuations, landfilling methods, nature of waste type and composition, and landfill design. Due to excessive rainfall during the wet season and high evaporation rates in tropical nations, such as Malaysia, landfill leachate is quickly produced.

Knowledge in improving the treatment for this landfill leachate management control is still an ongoing process and attracted significant attention from scientists, engineers, and technologists throughout the world. Various leachate treatment techniques were developed in many different ways, including those that incorporate biological, physical, and chemical processes and their combinations. Further improvements, especially on the optimisation for cost saving, are part of the continuing efforts.

The main problem with landfill leachate is the amount and level of variability that it exhibits [4]. In order to choose the best leachate treatment method, it is necessary to characterise the leachate and estimate its risk. In addition, treating the high concentration of NH_3-N in landfill leachate is a challenging process. The physicochemical method successfully eliminates heavy metals, inorganic macro-components, and refractory organic molecules from leachate. On the other hand, biological processes effectively remove dissolved organics and nutrients from leachates [5].

The age and biodegradability of the leachate limit the biological process for leachate treatment [6,7]. Physical-chemical techniques are normally necessary for lowering hazardous and refractory substances [8]. As a result, an integrated strategy combining biological with either pre- or post-physical-chemical processes is an effective choice that offers higher effluent quality [9]. The purpose of this paper is to review and summarise various physicochemical technologies in leachate treatment and compare their performances and limitations. There are many physicochemical treatments in the literature that provide effective techniques to deal with substantial organic content in leachate. The review also helps to better understand the suitable methods for specific types of leachate which may be applied in the field.

2. Landfilling

Landfilling stands among the main elements in a solid waste management strategy even after the implementation of the 3Rs: reduce, recover, recycle. This is due to the fact that, in most cases, not all waste can be recovered and recycled. Open dumping or regulated dumping are the two main methods used by most developing countries to dispose of their waste. These countries normally have financial constraints to apply costly treatment systems such as materials recovery facilities, waste-to-energy technology, etc. For the same reason, Malaysia currently relies primarily on landfilling as its main way of disposing of its MSW, and this is expected to still be the favoured option for the next 10 to 15 years to come. However, due to economic limitations, proper sanitary landfill concepts have not yet been fully implemented in the whole country. There are many old and improperly designed landfill sites that are still in operation to date, and some of them are almost reaching their end of life. The leachate is still being produced and must be properly controlled.

The category of landfill could be generally grouped into anaerobic, semi-aerobic, and aerobic. In developing countries, the anaerobic landfill is the most common, where the waste is commonly discarded and covered and, sometimes, left uncovered. Open dumping is still being practised, but the trend is now towards control tipping, and more countries are moving forward towards the sanitary landfill. The anaerobic landfills produce concentrated leachate, which is difficult to treat by a conventional method to up to the standard discharge limits. This type of landfill is further constrained by fire incidents and greenhouse gas emissions, which primarily contain methane (CH_4) and carbon dioxide (CO_2). Quite commonly, these gases are untapped and just released untreated in developing countries due to many limitations, especially the cost factor.

On the other hand, the Fukuoka technique, or a semi-aerobic method, was established at Fukuoka University over 20 years ago and was employed in numerous sites around Japan, China, Iran, and Malaysia. However, this method was not widely adopted by many other countries. Semi-aerobic landfilling was first used in Malaysia in 1988, and since then, the quality of the leachate has improved noticeably. The Fukuoka approach can be used in developing nations in a variety of situations for various goals, such as creating new landfill sites, improving existing ones, or effectively closing ones that were already constructed. Leachate collection pipes are built beneath the semi-aerobic landfill, as shown in Figure 1.

Leachate is removed by this conduit from a disposal location. Air from an open pit is extracted into this leachate collection pipe, which then moves into the waste body. In this manner, greenhouse gases such as methane and carbon dioxide are produced less, as the process encourages aerobic biodegradation or organic matter and allows early waste stabilisation [10]. Figure 2 illustrates a typical layout of a semi-aerobic landfill.

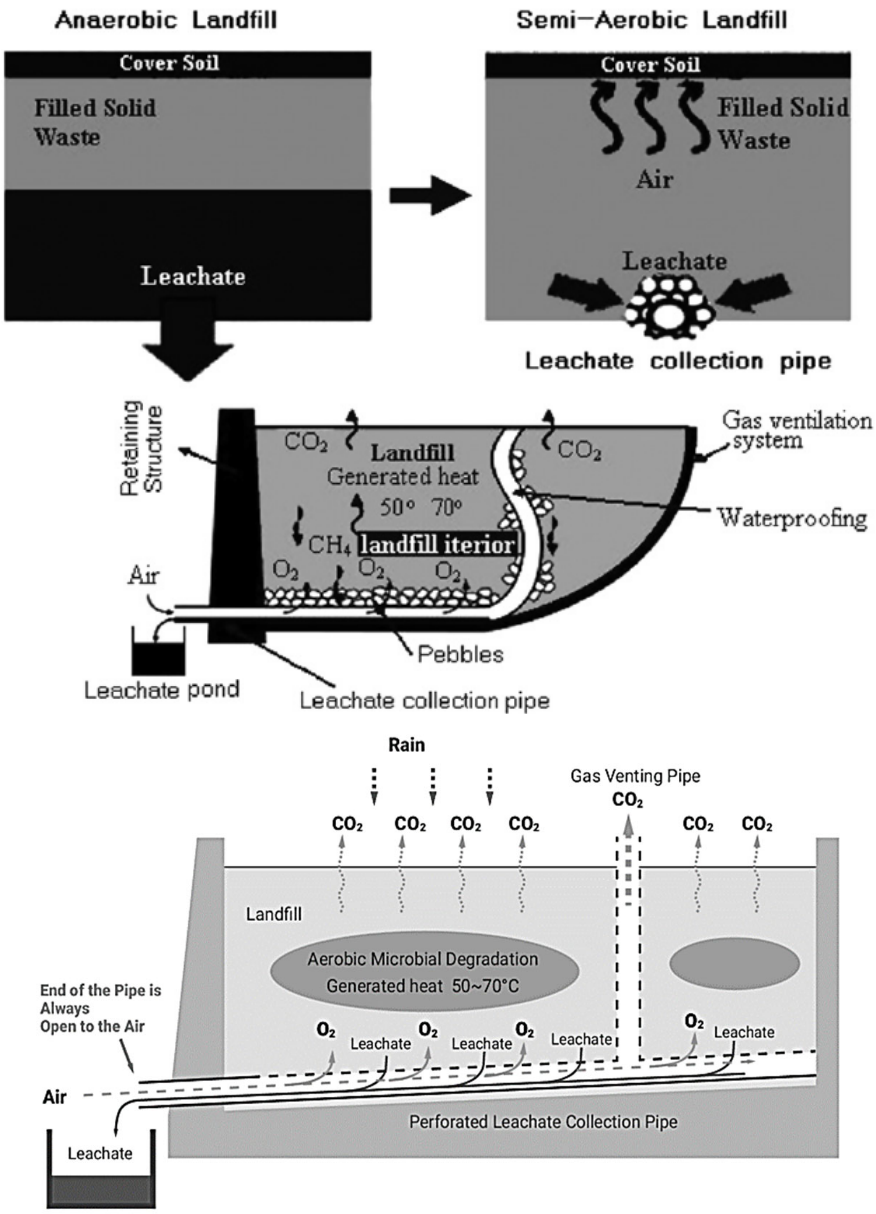

Figure 1. Conceptual landfill design using a semi-aerobic concept [11].

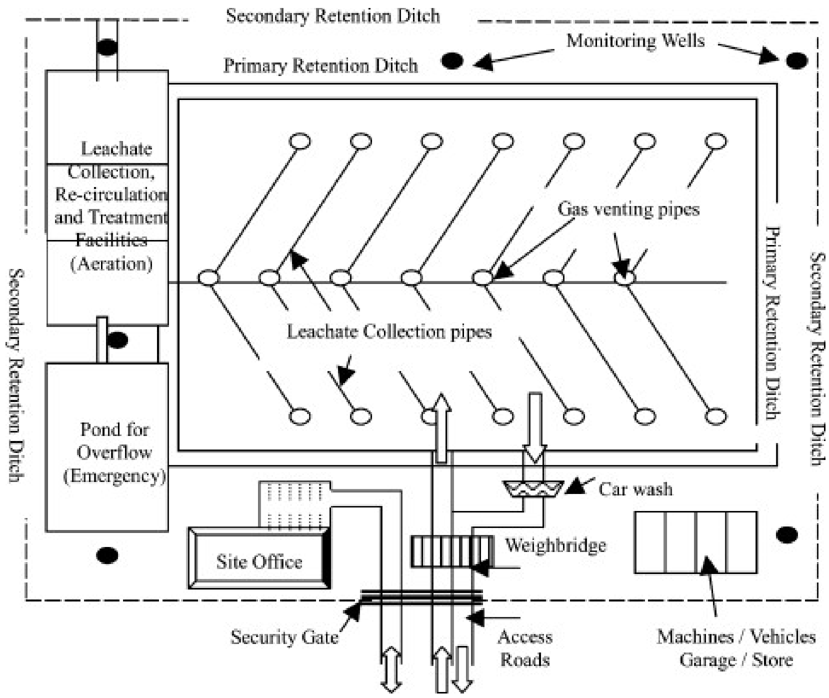

Figure 2. An illustration of a landfill that uses a semi-aerobic process [12].

Gas venting and leachate collecting pipelines are crucial components of a semi-aerobic landfill system. Figure 3 shows semi-aerobic landfill leachate pipes which provide air passage and remove leachates from the waste body through the natural convection process of cold air (outside) and hot air (within the waste body). This process mimics human blood veins [13]. Additionally, these pipelines have a number of benefits. Leachate, for instance, is evacuated more quickly than it would be in landfills without these pipelines. As a result, leachate fouling in waste materials is avoided and landfills are conveniently accessible to fresh air. Aerobic environments promote microbial activity and enhance waste decomposition. Due to their placement within rocks, collection pipelines are shielded both from clogging and operational harm. Leachate seepage is less likely because leachate is quickly drained, which lowers the pressure brought on by water on the ground [14].

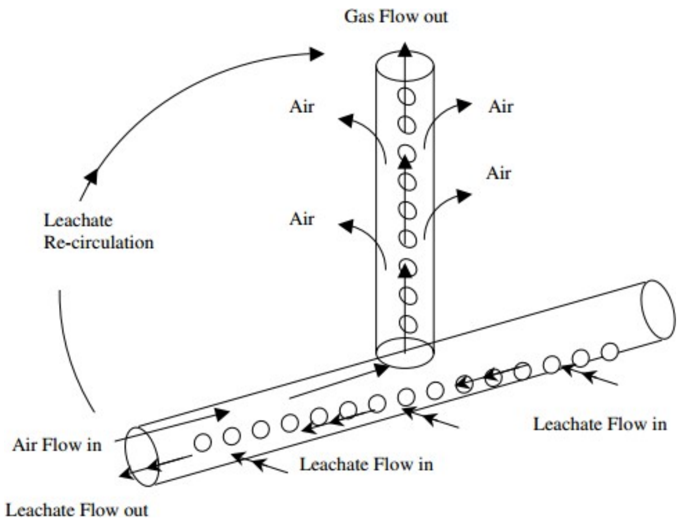

Figure 3. Leachate collection and gas ventilation pipes in a semi-aerobic landfill [12].

Landfills that employ a semi-aerobic concept are potentially sustainable, as they could offer various advantages over alternative solutions. When leachate flows through the pipes and is released from the sites, it lowers water pressure and prevents seepage. Garbage naturally allows fresh air to pass through it. Thus, leachate cleaning and waste stabilisation only take a short while. The amount of methane released decreases, despite an increase in carbon dioxide content. Semi-aerobic landfills also require straightforward technology, simple setup and operation, a small number of engineering protocols, equipment, and machinery, easier maintenance and operation, and inexpensive startup costs. Semi-aerobic landfills also contribute to a reduction in global warming by limiting the release of methane [13].

3. Characteristics of Landfill Leachate

Leachate is abundant in a wide variety of compounds (organic and inorganic), including humic and fulvic substances, heavy metals, fatty acids, and other potentially harmful compounds. Table 1 outlines the typical characteristics of landfill leachate. Numerous papers noted considerable variations in the components of the leachate. However, landfill age was used to identify three categories of leachates: fresh (under 5 years), transitional (5–10 years), and stabilised or old (over 10 years). Leachate quality can generally be controlled by a range of elements, such as the age of the site, rainfall, weather changes (which are seasonal), the nature of waste, and the waste properties. Fresh leachate is rich in organics and is highly biodegradable. NH_3-N is dominant in aged and stabilised landfill; it is normally non-biodegradable. Transitional landfill leachate has an intermediate quality between the young leachate and the mature leachate [15].

Table 1. Typical compositions of landfill leachate.

			Type of Landfill Leachate		
No.	**Parameter**	**Unit**	**Young (<5 Years)**	**Intermediate (5–10 Years)**	**Stabilised (>10 Years)**
1	pH		<6.5	6.5–7.5	>7.5
2	COD	mg/L	>10,000	4000–10,000	<4000
3	BOD_5/COD		0.5–1.0	0.1–0.5	<0.1
4	Organic compound		80% VFA [a]	5–30% VFA [a] + HFA [b] HFA [b]	HFA [b]
5	NH_3-N	mg/L	<400	NA [c]	>400
6	TOC/COD		<0.3	0.3–0.5	>0.5
7	Kjeldahl nitrogen	g/L	0.1–0.2	NA [c]	NA [c]
8	Heavy metals	mg/L	Low to medium	Low	Low
9	Biodegradability		Important	Medium	Low

Note: Source: [15]. [a] VFA is volatile fatty acid. [b] HFA is humic and fulvic acid. [c] NA is not available.

High colour intensity indicates that leachate consists significant content of organic substance, as colour is one of the important indicators of organic loading [16]. Concentrated COD, colour, and ammoniacal nitrogen (NH_3-N) were discovered during long-term landfill leachate monitoring in Malaysia. These high concentrations are regularly regarded by landfill operators as urgent problems that require proper attention.

4. Landfill Leachate Treatment

Among the biggest issues in managing landfill is determining how to deal with enormous and considerable amounts of leachate. A variety of techniques, including physicochemical, biological, and chemical processes, are normally required to remediate leachate effectively. These techniques typically involve numerous expensive and labour-intensive operations. Leachate treatments are difficult to perform owing to their hefty loading, complicated compounds, and flow that changes with the seasons [17]. The effectiveness of leachate treatment is generally improved with the right combination of treatment methods. Various combinations were reported [18]. Table 2 listed the options for treating leachate based on landfill age. A few of the typically employed methods to treat landfill leachate are shown in Figure 4. This is followed by Table 3, which provides different leachate treatment results according to landfill age. Cherni et al. [19], Mojiri et al. [7], and Teng et al. [5] reviewed the effectiveness of several leachate treatments from various literature sources. This is followed by Table 4, which presents the removal of leachate parameters by numerous applications by Matsufuji [20]. In Table 5, a summary of various advanced oxidation processes (AOPs) employed in processing leachate is presented as sourced and reviewed by Cherni et al. [19] and Teng et al. [5].

Table 2. Options for treating leachate based on landfill age.

Leachate Treatment	**Landfill Age (Years)**		
	Young (<5)	**Intermediate (5–10)**	**Mature (>10)**
Co-treatment with domestic wastewater	Good	Fair	Poor
Recycling	Good	Fair	Poor
Aerobic process (suspended growth)	Good	Fair	Poor
Aerobic process (fixed film)	Good	Fair	Poor
Anaerobic process (suspended growth)	Good	Fair	Poor
Anaerobic process (fixed film)	Good	Fair	Poor
Natural evaporation	Good	Good	Good
Coagulation/flocculation	Poor	Fair	Fair
Chemical precipitation	Poor	Fair	Poor
Carbon adsorption	Poor	Fair	Good
Oxidation	Poor	Fair	Fair
Air stripping	Poor	Fair	Fair
Ion exchange	Good	Good	Good
Microfiltration	Poor	-	-
Ultrafiltration	Poor	-	-
Nanofiltration	Good	Good	Good
Reverse osmosis	Good	Good	Good

Note: Adapted from reference [21].

Table 3. An overview of various technologies in treating leachate.

Coagulation Flocculation					
Parameter (Removals)	Turbidity (90%) NH_3–N (46.7%) COD (53.9%)	TP (47%) TOC (15%) NH_3–N (20%) TN (4%)	COD (61.9%) Colour (98.8%) SS (99.5%)	Organic Matter (22.57)	
Electrocoagulation (EC)					
Parameter (Removals)	(with Al electrodes) COD (70%) TN (24%) Colour (56%) Turbidity (60%)	(with Fe electrodes) COD (68%) TN (15%) Colour (28%) Turbidity (16%)	COD (60%) NH_3–N (37%) Colour (94%) Turbidity (88%) SS (89%)	heavy metals Cr (51%) As (59%) Cd (71%) Zn (72%) Ba ((95%) Pb (>99%)	
Adsorption					
Parameter (Removals)	COD (77.3%) Colour (82.5%)	COD (93.6%) NH_3–N (84.8%)	Colour (100%) COD (~80%) $NH3^+$-N (100%)	COD (36%) NH_3–N (99%) Cl (18%)	COD (51.0%) NH_3–N (32.8%) Cl (66.0%) Br (81.0%) Cu (97.1%)

Note: Adapted from: [5,7,19].

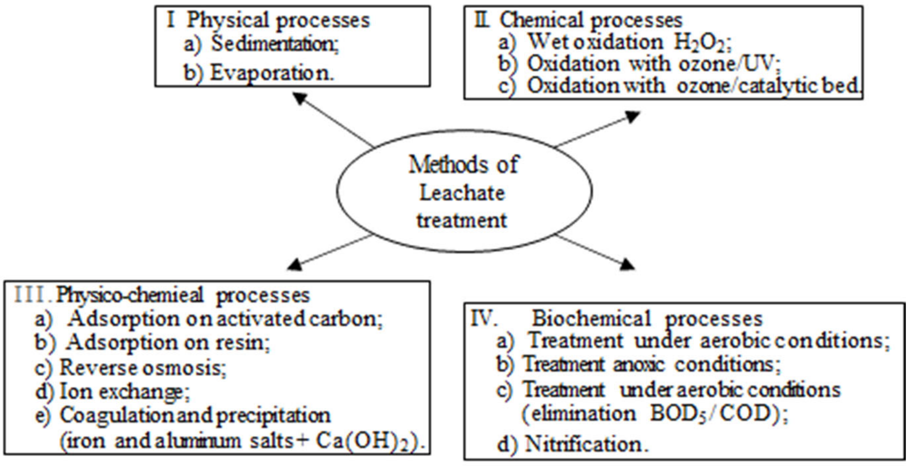

Figure 4. Techniques frequently employed at municipal landfills to treat leachate [3].

Table 4. Removal of leachate parameters by various applications performance [20].

Treatment Method	**Leachate Parameters**					
	BOD	**COD**	**SS**	**NH_3-N**	**Colour**	**Heavy Metals**
Activated Sludge Process	▲	•	Ø	Ø	Ø	Ø
Contact Aeration Process	▲	•	Ø	Ø	Ø	Ø
Rotary Biodisk Conductor Process	▲	•	Ø	Ø	Ø	Ø
Biological Trickling Process	▲	•	▲	Ø	Ø	Ø
Biological Nitrogen	▲	•	Ø	▲	Ø	Ø
Flocculation-Sedimentation	•	▲	▲	Ø	▲	•
Sand filtration	Ø	Ø	▲	×	•	×
Activated Carbon (Adsorption)	▲	▲	•	Ø	▲	•
Chemical Oxidation	×	•	×	×	▲	×

Notes: High (▲) Medium (•) Low (Ø).

Table 4 demonstrates the effectiveness of biological processes in treating leachates from newly constructed landfills (less than five years old). Biological treatment is normally employed when the leachate is biodegradable ($BOD_5/COD > 0.3$). As shown, biological treatment is the most appropriate method for fresh leachates containing high concentrations of organic material (>10,000 mg/L). For mature and stabilised leachates which are in the methanogenic phase (>10 years), the biodegradability ratio (BOD_5/COD) is normally less than 0.1, and it is hard to be biodegraded. It is normally rich in humic and fulvic acids [21]. For leachates with elevated NH_3-N levels and limited biodegradability, a physical chemical approach is the best option.

Malaysia uses a variety of leachate treatment techniques. Figure 5 illustrates one of the promising leachate treatment systems in Malaysia for a moderate site which receives 1500 tons of MSW per day.

Table 5. Summary of advanced oxidation processes (AOPs) in treating leachate.

AOP	Removal Efficiency		AOP	Removal Efficiency	
	Parameters	**Removal (%)**		**Parameters**	**Removal (%)**
Fenton	TOC COD	68.9 69.6	Ozone (O_3)	Colour COD Ammonia	100 88 79
	COD	88.6		COD Colour	70 100
	COD	70		COD Colour	16.5 40.5
	COD UV_{254} Colour	58.70 85.69 88.30		Humic Acid Fulvic Acid	88 83.3
	COD	97.8 3		COD	43
	COD BOD_5	48 30		COD TOC BOD_5	65 62 36
	COD TOC	97.83 74.24		COD UV_{254}	46 51
	total organic carbon, total inorganic carbon total nitrogen, colour	88.7 100 96.5 98.2		Colour UV	~90 ~70
TiO_2 Photocatalysis	COD Colour	58 36	Electro-oxidation	COD TOC	68 40.6
	COD TOC	67 82.5		COD	80
	COD	84		TOC Ammonium nitrogen	40 99
Ferrosonication (FS)	COD BOD_5	46 33	W-doped TiO_2	COD	46
Heterogeneous catalytic ozonation (O_3/TiO_2)	COD NTU BOD_5	24 94 98	Heterogeneous catalytic ozonation (O_3/ZnO)	COD NTU BOD_5	33 95 98

Note: Adapted from: [5,19].

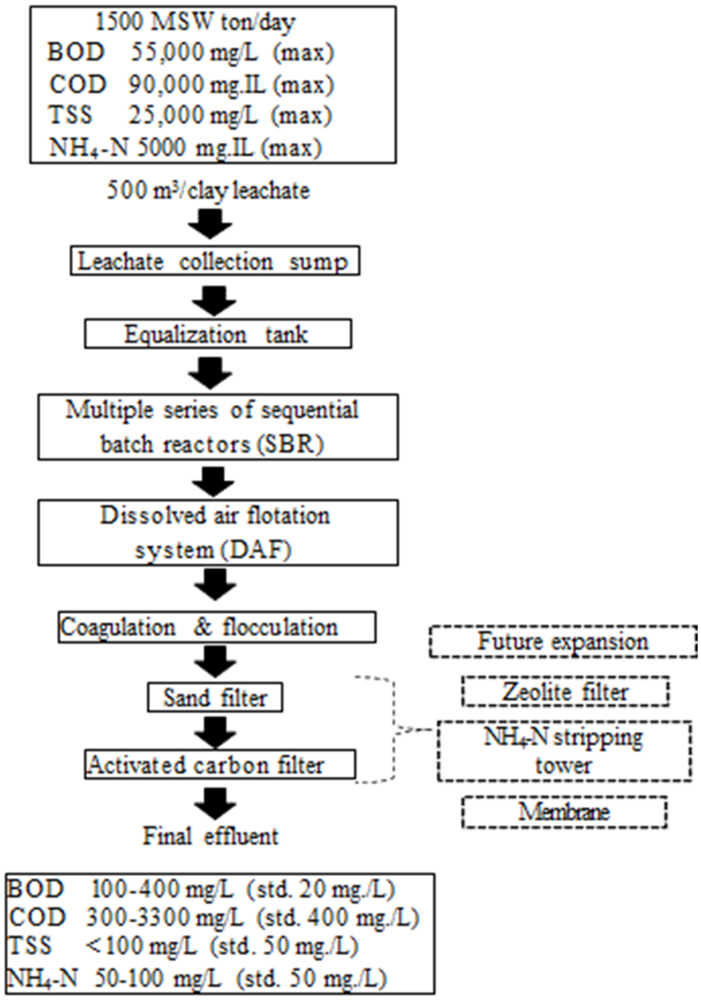

Figure 5. A typical landfill leachate treatment system in Malaysia for a moderate site landfill site (1500 tons/day) [22].

5. Physicochemical Treatment for Landfill Leachate

Biological processes have limitations in treating recalcitrant and non-biodegradable compounds with a BOD_5 to COD ratio of less than 0.5. Leachate treatment using physicochemical methods is often more efficient financially and quicker to complete for this kind of leachate [23]. This physicochemical approach can be used to treat old leachate that has highly elevated levels of COD and NH_3-N, low BOD, and good oxidation-reduction ability. The most popular physicochemical processes are coagulation-flocculation, adsorption, chemical precipitation, reverse osmosis, ammonia stripping, and oxidation [24]. Figure 6 illustrates the criteria that should be considered when selecting a treatment for landfill leachate [21].

The necessity to boost the efficacy of biological systems led to the development of physicochemical procedures. They are, therefore, frequently used following a biological pre-treatment. This technique functions by changing the chemical makeup of specific compounds or the physical components that can trap or remove pollutants. Table 6 lists some of these technologies' benefits and drawbacks [24].

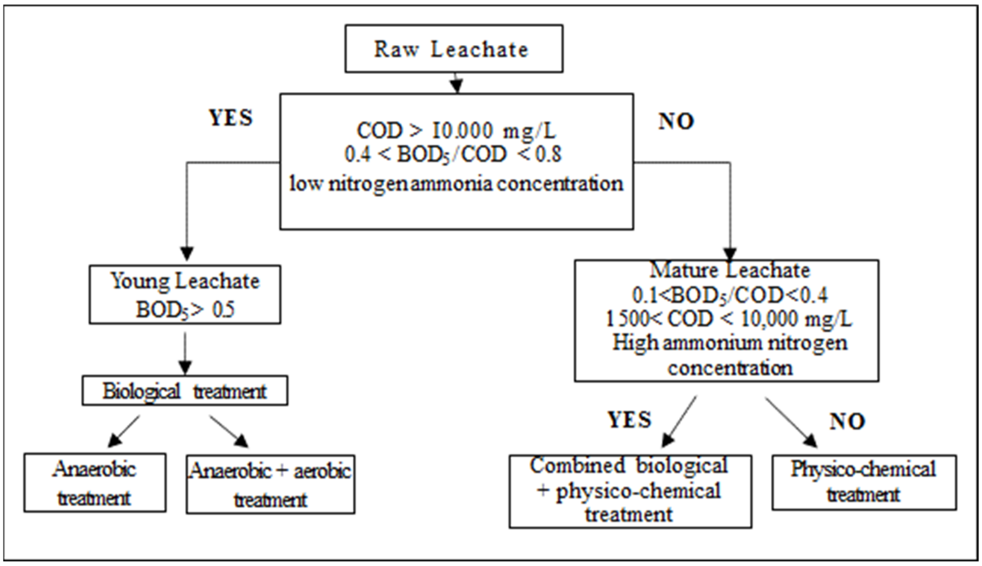

Figure 6. A typical leachate treatment selection protocol. Source [21].

Table 6. Leachate physicochemical treatment system benefits and drawbacks.

Physicochemical	Advantages	Disadvantages	Observations
Coagulation and Flocculation	Effective at removing suspended particles, humic acids, heavy metals, and organic matter.	Owing to the expense of inputs and the handling of the created chemical sludge, the system's functioning requires very high coagulant concentrations, making it economically impracticable to implement this technology on a large scale.	For some membrane systems, this technology serves as a pre-treatment. Some membrane systems appear to use this technique as pre-treatment.
PACT (Powdered Activated Carbon Treatment)	Removes some poisons, chlorine, phenols, ammoniacal nitrogen, colour, odour, and taste. Safeguards the process against BOD and organic toxin shock loads by stabilising it. It is simple to use, operate, and maintain and has inexpensive installation costs. Pre-treatment technology is used with several membrane systems.	High operating expenses with on-site regenerating or coal deployment, as well as outputs with high potential pollutants	Aeration, biological oxidation, and physical adsorption happen at the same time as coal is supplied directly to the reactor.
Advanced Chemical Oxidation	It divides these high molecular weight molecules, which increases their treatability by making them more receptive to microorganisms in biological reactors and partially eliminates recalcitrant organic material and refractory chemicals.	Due to the complexity of the operation and the high cost of operation, such as energy and the value of the inputs necessary in significant doses, a competent technical operator is required.	The most common oxidative technology is ozonisation.
Evaporation	Up to 95% reduction in leachate volume.	Polluting gases are released, and it costs a lot of energy-60 kg of gasoline are required to burn 1 m^3 of leachate. An output of dry sludge equal to around 5% of the entire volume is produced.	The option that is most frequently used is the landfill's own biogas being captured and burned.

Note: Adapted from: [19].

5.1. Ammoniacal Nitrogen (NH_3-N) Reduction by Air Stripping

The content level of ammoniacal nitrogen (NH_3-N) in landfill leachate level can be decreased or removed effectively using the air stripping technique. Basically, this physical mechanism is effective when the pH is 11. This ammonia loss is caused by desorption through the water surface, and the process is influenced by temperature. The stripping techniques are significantly influenced by the leachate quality and the treatment reactor layout. By raising the flow rate of the incoming air (air: waste flow rate) and employing smaller bubble diffusers, ammonia elimination can be increased [25]. The primary disadvantage of air stripping is the environmental pollution brought on by the release of NH_3 into the atmosphere. Consequently, further exhaust gas treatment is required. However, the NH_3-N becomes gas at this pH, and this gas, combined with the acidic solution, will produce ammonium salts that can be utilised as fertiliser in the mineral form [19]. Another key drawback of this technique is the limited effectiveness of organic matter removal, even though that air stripping is excellent at removing NH_3-N. Despite its ability to remove 90% of the ammonia at 20 °C, air stripping alone normally could not meet effluent discharge restrictions. As a result, the subsequent nitrification and denitrification process via air stripping can probably meet the effluent discharge guidelines. Furthermore, despite the removal of COD and ammonia, air stripping was found to increase toxicity in several laboratory tests. However, air stripping was discovered to be the most economical alternative approach for high ammonia removal when compared to other processes such as membrane filtration [26].

A number of operational conditions, including pH, initial NH_3-N level, hydraulic gas-to-liquid ratio (G/L), loading rate, and recirculation period [27], influence the air stripping system. Their study showed that, regardless of changes in the G/L or hydraulic loading rate (HLR), increasing pH from 9 to 12 resulted in a considerable improvement in ammonia removal efficiency, with the maximum ammonia stripping achieved at pH 12. As the G/L ratio rose, the removal efficacy increased by up to 56% for both HLRs of 57.6 and 172.8 m^3/m^2/day. Under the following conditions: HLR of 172.8 m^3/m^2/day, pH 12, G/L of 728, and liquid recirculation, 99% of its ammonia (2520 mg/L) was stripped within three hours. The ammonia concentration in the final sample was 25.2 mg/L, which is almost as good as the allowable discharge limit.

Leite et al. [28] investigated and characterised the NH_3-N stripping method at open horizontal flow reactors. The study involved the superficial load in three different phases phase 1 (650 kg N-NH_4^+ $ha^{-1}.day^{-1}$), phase 2 (750 kg N-NH_4^+ $ha^{-1}.day^{-1}$), and phase 3 (850 kg N-NH_4^+ $ha^{-1}.day^{-1}$). The procedure demonstrated a removal efficiency of 99.0%, 99.3%, and 99.5% in the first, second, and third phases, respectively. In addition, phases 1 and 2 had removal efficiencies of 69.2%, 40.12%, and 29.23%, respectively, for organic matter reported in terms of total COD. Following a series of tests, the researchers concluded and demonstrated that the effectiveness of ammonia removal was directly connected with the surface load, but the effectiveness of carbonaceous material removal was correlated with the amount of organic matter applied in the influent.

5.2. Coagulation-Flocculation Process

Prior to a biological process, coagulation-flocculation is one of the several physico-chemical treatments that is generally employed in the preparation of stabilised and matured landfill leachates [19]. Generally, the physical-chemical through a coagulation-flocculation process involves the destabilisation of small particles (colloids) in wastewater to create flocs that could be easily precipitated. In an effort to destabilise the colloidal particles, various coagulants react differently with colloidal particles [29]. Coagulants commonly occur in a variety of forms. The most often used coagulants are chemical-based ones, such as alum and ferric salts. When trace metal salts such as ferrous sulphate, aluminium sulphate, and ferric chloride are added during the coagulation-flocculation stage, high valence cations are formed in the solution, which lowers the zeta potential values. Ferric ion salts generally outperform aluminium salts, primarily due to their insoluble nature over a wider pH range [19]. These destabilising occurrences are caused by a variety of mechanisms, such as charge neutralisation, trapping, adsorption, and complexation with the metal ions of the coagulant to create insoluble aggregates [30]. The dosage and types of coagulant/flocculant, as well as the experimental settings such as pH, time, and temperature, have a considerable effect on the coagulation efficacy [31]. In addition, the efficacy of the coagulation method, which removes organic matter and phenolic compounds from wastewater, is also influenced by the mixing conditions and the characteristics of phenolic compounds, such as particle size, charge, and hydrophobicity. The coagulation-flocculation approach was used successfully to eliminate non-biodegradable organics, suspended solids, colloidal particulates, colour, turbidity, and heavy metals, depending on the pollutant and types of coagulant and flocculant [30].

Leachate treatment via the coagulation-flocculation technique was the subject of a great deal of research. Djeffal et al. [32] investigated the efficacy of the coagulation-flocculation technique for the purification of leachate from the Souk-Ahras City Technical Centre landfill in Algeria. Three distinct coagulants; ferric chloride, aluminium sulphate, and alum, as well as two agitation techniques; mechanical and ultrasound, were applied in the study to optimise the operational conditions. With a 15% coagulant dosage, 250 rpm stirring rate, and a response time of roughly 15 min for ferric chloride, a considerable drop in turbidity (99.4%) was made possible. The turbidity was reduced by 98.9% and 98.6%, using aluminium sulphate and alum, respectively, with the other two coagulants having an optimal coagulant-to-leachate volume ratio of one. The results of the bacteriological tests also showed a lack of *Streptococci*, total germs, and faecal coliforms. Furthermore, when a 37 kHz ultrasonic waves frequency of 30 W power was used to treat the leachate, it was discovered that the turbidity of the supernatants greatly decreased.

Mohd-Salleh et al. [33] employed polyaluminium chloride (PAC) in treating leachate in various operating settings (variable dosage and pH). The objectives of the study were to identify the best coagulant dosage in a range of dosages (2250–4500 mg/L), as well as to examine the best pH (pH 3–10). This was carried out through different sets of jar tests to assess the influence of five different leachate parameters, including suspended particles, NH_3-N, COD, and heavy metals, on the removal efficacy. They concluded that the ideal PAC dosage and sample pH was 3750 mg/L and pH 7, respectively. Reductions of 95%, 53%, 97%, and 79% in the suspended particles, COD, Fe, and Cr, respectively, were achieved at this ideal dose and pH.

5.3. Adsorption

Molecules from a normally liquid medium (the adsorbate) are pulled to and maintained on the surface of the other, frequently solid medium (adsorbent) as a surface phenomenon (adsorption). A large surface area of the adsorbent is needed to boost the treatment's efficiency because the process takes place on its surface. The adsorbate can attach to the surface of the adsorbent due to the specific properties of its surface. Because adsorption includes a surface mechanism, the adsorbent surface area is important, and an adsorbent with a greater surface area and increased porosity offers the adsorbate more interaction sites [34]. It is also conceivable for a reversible phenomenon known as desorption to occur when adsorption takes place under specific circumstances. Adsorbates are transported back to the liquid phase during desorption after being liberated from the adsorbent's surface. Figure 7 illustrates the fundamental idea of adsorption [35].

Physical adsorption, also known as physisorption, and chemical adsorption, sometimes referred to as chemisorption, are the two categories into which adsorption can be separated. In essence, the bonding between the two forms of adsorption varies. Van der Waals forces of attraction among the adsorbent and adsorbate produced physical bonding in the process of physisorption; meanwhile, adsorbate and adsorbent in chemisorption were attracted to one another with force comparable to chemical bonding [36].

Due to the massive production of activated carbon (AC) adsorbents, the commonest form of adsorbent, the application of the adsorption technique substantially expanded over the years. Because of its highly porous surface area, thermostability, and exceptional ability, a wide variety of organic and inorganic contaminants dissolved in aqueous media can be removed by AC with remarkable efficiency. The pollutants in AC in columns typically reduce COD levels more efficiently than chemical treatment methods. Because of this, there is now a much higher chance that the high quantities of organic chemicals in leachate may be removed via the adsorption technique [37].

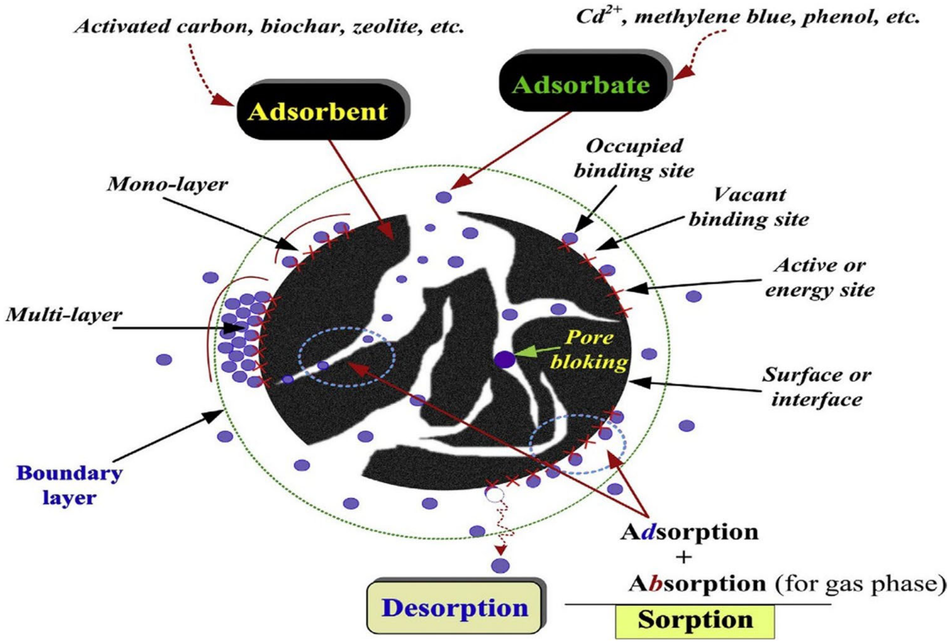

Figure 7. The adsorption fundamentals. Source: [35].

Contemporary porous materials were proposed for improved adsorption capacities and reduced environmental effects [38]. These materials are available in a diverse range of chemical configurations, surface finishes, and geometries. Adsorbents are often constructed from a variety of substances, such as natural substances, enhanced natural substances (such as activated carbon), synthetic substances (such as zeolites and resins), industrial and agricultural wastes and by-products, biological adsorbents, and others [39]. There were numerous different categories and types of adsorbents reported in the literature. Some of them are simplified graphically in Figure 8 [40].

In earlier investigations, many researchers applied and utilised a variety of media, including chitosan, activated carbon, zeolite, clay, and others, to remediate leachate [7,41]. Rohers et al. [42] published their work on the effectiveness of activated carbon in leachate treatment via a column study. Sand filters and activated carbon columns were used as an option for the physical-chemical leachate pre-treatment in their work. The results showed that COD, BOD_5, colour, and NH_3-N decreased by up to 74%, 47%, 93.4%, and 90%, respectively. Their work was based on a wider range of the BOD_5/COD proportion from 0.3 to 0.9. Additionally, the NH_3-N concentration was decreased by 85.37% using an activated carbon post-filtration column. The post-treatment also led to significant heavy metal reductions (60–96%).

Other naturally occurring minerals, including clays and zeolite, are also good adsorbents to complement AC in treating leachate. Natural minerals with excellent adsorption and ion exchange abilities, such as zeolites (clinoptilolite), were proposed for use as adsorbents due to their low cost [33,43]. Many workers researched how natural zeolite affected ammonium ions (NH_4-N) remediation from leachate [44]. The impacts of a number of variables, including reaction time (T), pH, and zeolite concentration (ZC), were examined in a batch process to optimise the process. The first step was to investigate the effects of pH at different pH ranges (pH 5 to 9). The influence of the ZC in the pH-optimal range of 10–200 g/L was next assessed. According to the results, raising the ZC from 10 to 80 g/L improved the elimination of NH_4-N. When the ZC concentration was raised from 80 to 200 g/L, the performance unfortunately decreased due to the overdosage phenomenon. The studies' findings demonstrated that a pH of 7, a ZC of 80 g/L, and a reaction time of 30 min were required for removing NH_4-N (44.49%). The work demonstrated that the clinoptilolite could be used to effectively and economically extract ammonium ions from landfill leachate [44].

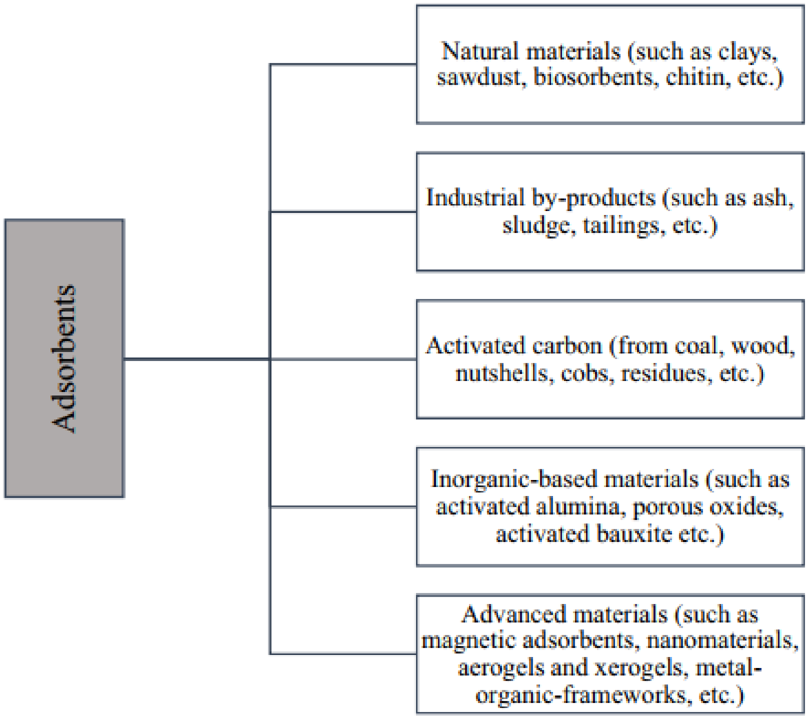

Figure 8. Classification of adsorbents. Source: [40].

The efficacy of raw zeolite and heated activated zeolite to possibly reduce COD, ammoniacal nitrogen, and colour in leachate was investigated by Aziz et al. [34] in 2020. The zeolite applied in the study was heated for three hours at various temperatures for activation. The optimal dosage was determined to be 10 g of raw zeolite, which reduced 53.1% NH_3-N, 22.5% COD, and 46% colour. As much as 24.3% of COD and 73.8% of colour were reduced at pH 4. At the optimal pH of pH 7, roughly 55.8% of the NH_3-N was decreased. The best dosage was 10 g of heated activated zeolite at 150 °C, and this temperature led to reductions in 45.1% NH_3-N, 11.8% COD, and 43.7% colour. The heating temperature of 150 °C exhibited the best performance and was cheaper, which showed potential to be upscaled in the field. Additionally, the zeolite's capacity was improved and increased from 41.30 cmol/kg to 181.90 cmol/kg by heating.

5.4. Integrated Treatment

It is quite common to combine physicochemical and other treatment methods in treating leachate to adhere to the acceptable threshold for effluent release. Combining treatments was shown to be more economical and affordable for treating mature leachate due to their capacity to synergistically enhance the benefits of each of the methods used [45]. The combination of two or more biological, physiochemical, and biological-physical-chemical processes are among the common workable hybrid approach in the treatment. The hybrid approach combines many technologies to produce a product that is better for the environment and could be used at a lower cost at once, as no additional post-treatment is necessary [46]. Many landfill leachate sites already combine a biological treatment with an adsorption pre-treatment [47]. Some of the common hybrid/combined approaches that demonstrated effective treatment of landfill leachate are presented here.

Mohajeri et al. [48] investigated the sequencing batch reactor (SBR) mixed with powder of sawdust-enriched bentonite as an adsorbent in removing organic chemicals from established landfills. Based on their pH values, the sawdust was examined at neutral, alkaline, and acidic conditions. At the ideal aeration speed of 7.5 L/min and reaction time of 22 h, SBR-augmented bentonite treatment eliminated COD and NH_3-N by 99.28% and 95.41%, respectively. It was a notable success that, even with the reaction time decreased to two hours, the removal of both contaminants in the existence of sawdust only decreased to 17%.

de Oliveira et al. [49] reported their findings in remediating landfill leachate employing a combination of a filter-press reactor, a coagulation-flocculation (alum) process, and electrochemical approaches with a boron-doped diamond electrode. pH 6.0 and 20 mL/L $Al_2(SO_4)_3$ (50 g/L) was proven to be the most favourable condition for the coagulation-flocculation process. Three distinct coagulants, ferric chloride, aluminium sulphate, and alum, as well as two agitation techniques, ultrasound and mechanical, were used in the study to optimise the operational conditions. This process used up to 40% less energy to remove the organic load while keeping a similar efficient mineralisation rate (>90% COD reduction). By the end of the electrolysis process, colour, turbidity, and NH_3-N were totally removed.

El Mrabet et al. [50] investigated the application of the Fenton method in conjunction with adsorption onto naturally occurring bentonite clay in a landfill leachate treatment. The optimum Fenton conditions occurred at 2000 mg/L of Fe^{2+}, 2500 mg/L of H_2O_2, and pH 3, which exhibited 92% and 73% reductions in colour and COD, respectively. The pre-treated leachate was then passed through the naturally occurring bentonite clay. The impacts of a number of factors, including pH, reaction time, adsorbent dosage, and temperature, on the adsorption effectiveness were examined. As much as 84% of the total COD and 98% of the colour were removed by the integration of the Fenton and adsorption processes (bentonite dosage: 3 g/L; pH 5; contact time: 5 h; temperature = 35 °C).

Table 7 summarises various combinations in treating landfill leachate as sourced and reviewed by Cherni et al. [19] and Mojiri et al. [7]

Table 7. Combinations of landfill leachate treatments from the literature.

Combination Treatment Category	**Removal Efficiency**		**Combination Treatment Category**	**Removal Efficiency**	
	Parameters	**Removal (%)**		**Parameters**	**Removal (%)**
Advanced oxidation process/coagulation/adsorption	COD As Fe P	94 87 96 86	Bioreactor/coagulation	Colour COD Ammonia TSS	85.8 84.8 94.2 91.8
Advanced oxidation process/adsorption	Ammonia COD Colour HA (ABS_{254})	94.5 95.1 95.0 97.9	Bioreactor/membrane	COD Fe Zn	95 71 74
Advanced oxidation process/adsorption (ion-exchange)	Ammonia Nitrite Nitrate Colour Turbidity COD	90 100 98 98 98 74	Advanced oxidation process/coagulation	COD Colour HA (UV_{254})	68 97 83
Electrodissolution/advanced oxidation process/chemical flocculation	COD Colour Turbidity	85 96 76		COD HA	90.2 93.7
				COD	91

Note: Adapted from: [7,19].

6. Challenges

1. Treating leachate is tough and demanding owing to its complex compounds, which involve large differences in its volumetric chemical compositions. Selecting an acceptable, cost-effective, and efficient combination procedure is a demanding undertaking.
2. Leachate normally varies in terms of loading due to large fluctuations in water quantity and quality. This is because it is greatly influenced by the amount of waste disposed of daily, season, and weather conditions, which make it difficult to choose and run an effective treatment method and consistent performance.
3. Treatment of leachate depends on its composition. As leachate properties differ, treatment methods for leachate A might not work well for leachate B. Therefore, a treatability study is highly recommended. Experiences and performances of an existing leachate treatment plant will complement this treatability study.
4. It is also not straightforward to determine an appropriate and the best combination of available technologies and how to combine them to achieve a steady operation.
5. Budget restrictions in developing countries make it challenging to establish and maintain an effective treatment system.
6. Treating NH_3-N and total nitrogen is a challenging task. Usually, a nitrification-denitrification system or an ammonia stripping plant is required, although they are a bit costly. Zeolite filters, however, recently became a promising method as an alternative in removing NH_3-N.
7. In addition, some leachate treatment facilities frequently employ post-treatment steps to polish the treated effluent. Nanofiltration and reverse osmosis were employed in some sites to meet discharge limits; however, this is costly and may not be widely affordable in developing countries.
8. There is a limitation of technical knowledge in underdeveloped countries on the management and operation of treatment facilities.

7. Conclusions

Leachate contamination is an increasing threat to the environment and human health, particularly in developing countries. Leachate not only affects underground aquifers and the Earth's ecosystem, but it also releases toxic pollutants and greenhouse gases into the atmosphere. As a result, mitigation of these negative effects is necessary; this requires a cost-effective, sustainable approach and environmentally friendly leachate treatment facilities.

In spite of the presence of different approaches in the treatment, no distinct and single method is normally sufficient, efficient, or economical enough to meet the requirements of effective standards. Further research is still ongoing to meet the demand, especially for developing countries.

In multiple-stage treatment systems, current trends involve a combination of biological, chemical, and physicochemical processes. Chosen techniques depend on many technical factors, which should be properly assessed and examined because one technique may not be adaptable to all situations.

The review assessed numerous leachate treatment technologies, their efficacy, and the benefits and limitations to the environment. Subsequently, it is necessary to research and develop an innovative technology which can optimise the performance of the treatment at an affordable cost, especially in reducing energy and chemical usage.

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