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Review

State of the Art in Anaerobic Treatment of Landfill Leachate: A Review on Integrated System, Additive Substances, and Machine Learning Application

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Abstract: Leachates from landfills are highly polluted with a considerable content of organic and inorganic pollutants which pose severe deterioration to environment including soil, groundwater, surface water and air. Several mitigative measures have been applied for effective management of leachate such as biological treatment, engineering device control leachate migration, physical/chemical treatment, and membrane technology. Among the alternatives, anaerobic digestion (AD) is promising, with effective removal of pollutants and high potential for renewable energy production and nutrient recovery. Landfill leachate (LFL) is an excellent source as a substrate in an AD system, with its high content of organic matters. The advantages and disadvantages of AD of LFL were extensively discussed in this review in terms of its potential as a co-substrate, pre-treatment application, and the types and design parameters of the digester. The review critically evaluated the previous studies on leachate treatment using an AD system as well as potential factors which can enhance the treatment efficiency, including the application of an integrated system, additive substances as well as potential inhibition factors. Pre-treatment methods have the potential to meet desired effluent quality of LFL before discharging into receiving bodies. The review also highlighted the application of kinetic modelling and machine learning practices, along with the potential of energy generation in AD of LFL. Additionally, the review explored the various strategies, and recent advances in the anaerobic treatment of LFL, which suggested that there is a requirement to further improve the system, configuration and functioning as a precursor in selecting suitable integrated LFL-treatment technology.

Keywords: leachate treatment; waste to bioenergy; leachate pollution; municipal solid waste; kinetics model

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Citation: Zamrisham, N.A.F.; Wahab, A.M.A.; Zainal, A.; Karadag, D.; Bhutada, D.; Suhartini, S.; Musa, M.A.; Idrus, S. State of the Art in Anaerobic Treatment of Landfill Leachate: A Review on Integrated System, Additive Substances, and Machine Learning Application. *Water* **2023**, *15*, 1303. https://doi.org/10.3390/w15071303

Academic Editors: Giovanni Esposito and Antonio Panico

Received: 10 February 2023
Revised: 17 March 2023
Accepted: 22 March 2023
Published: 25 March 2023

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1. Introduction

Leachate from landfills is considered a major concern to communities as it contains hazardous substances. LFL contains high concentrations of organic pollutants, salts, ammonia, nitrogen, and heavy metals, as well as xenobiotic organic materials such as phthalates [1,2]. Improper treatment and disposal of LFL pollutes surface water, degrades ecosystems, and negatively affects public health [3,4]. Additionally, LFL might percolate through the soil and contaminate ground water, with negative impacts on potential drinking water resources [5,6]. Inadequate disposal or treatment of the LFL is hazardous, and will lead to environmental pollution and social problems [7,8].

LFL contains heavy metals such as copper (Cu), nickel (Ni), chromium (Cr), zinc (Zn), and cadmium (Cd) that come from industrial and commercial waste [2,9,10]. Cd (group 1), Ni (group 1), Cr (group 1), and lead (Pb) (group 2B) are all categorized as potentially carcinogenic metals, while Cu, Zn, manganese (Mn), and aluminium (Al) are declared as non-carcinogenic metals [11]. Improper management or treatment can pollute water bodies and cause a severe impact on aquatic and human life. The accumulation of heavy metals in the human body creates serious health problems such as damage to the nervous system, headaches, coughing, depression, and kidney infection [9,11] Figure 1 depicts the adverse effects of heavy metals, which are present abundantly in LFL content. According to the Environmental Protection Agency (EPA), Environmental Protection Department (EDP), and Pollution Control Department (PCD), the range standard for effluent discharge for Cr (VI) is 0.05–2.0, Zn (II) is 0.6–5.0, Cu (II) is 0.05–4.0, Cd (II) is 0.001–0.2, and Ni (II) is 0.10–4.0 mg/L.

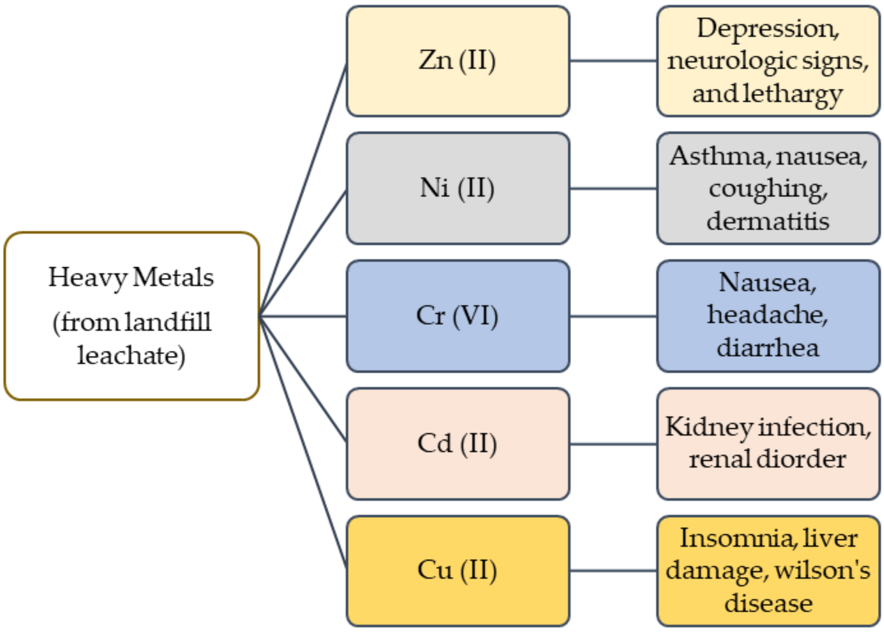

Figure 1. Effect of heavy metals to human health.

Impractical solid waste management (SWM) results in the production of LFL worldwide. LFL is formed when rainwater and moisture accumulate within a waste deposit, resulting in a highly polluted, dark-coloured, and odorous liquid [10]. LFL has emerged as one of the most pressing issues in SWM, requiring attention globally, primarily in treating young leachate since it contains higher chemical oxygen demand (COD) and biological oxygen demand (BOD) compared to intermediate and old LFL. Bove et al. [2] mentioned that, based on the European Waste List, the leachate can be classified as hazardous or non-hazardous waste. Hazardous leachates must undergo the preliminary physicochemical treatments before continuing with biological treatment (i.e., aerobic or anaerobic treatment), while non-hazardous leachates need to undergo the necessary analyses before performing the direct treatment. According to Bove et al. [2], Ahmad et al. [12], and Nawaz et al. [13], the properties of LFL differ according to age (young, intermediate, and old) and the waste composition of the landfill deposit, as presented in Table 1. The BOD/COD ratio decreases proportionally with increasing LFL age as biopolymers degraded [2]. In addition, the older the age of LFL, the higher the pH level, which in turn impacts the treatment efficiency. Renou et al. [14] reported that COD removal for old LFL was 75% (anaerobic sequencing batch reactor), 88% (up-flow anaerobic sludge blanket reactor), and 75% (hybrid bed filter). The COD removal for AD of intermediate LFL was slightly lower in an up-flow anaerobic sludge blanket reactor, in a range of 45–71%.

The generation of leachate has risen as the population has grown, which indicates the urgent need for efficient treatment. Figure 2 depicts the trends in the research on the treatment of LFL and anaerobic system applications published between 2005–2022.

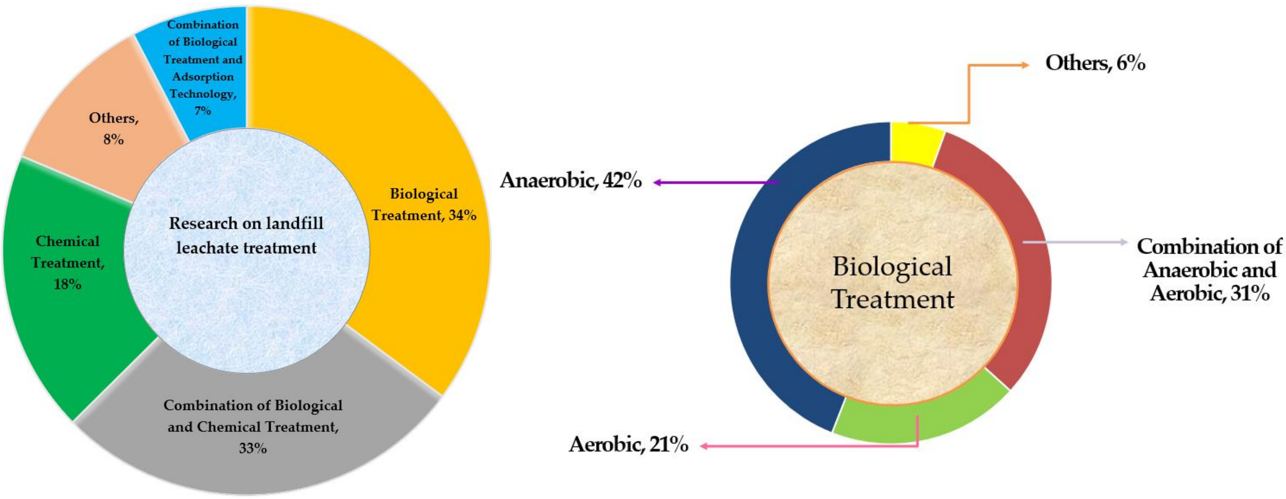

Figure 2. Research on the LFL treatment and anaerobic system application published between 2005–2022.

Most of the previous research has been conducted to identify appropriate methods for the treatment of LFL, as shown in Tables 2–4, which include biological processes (aerobic and anaerobic treatment), leachate transfer (recycling and techniques for the combined treatment of LFL with sewage), and physical/chemical treatments (adsorption, chemical precipitation, chemical oxidation, sedimentation, air stripping, and coagulation–flocculation (CF)) [2,13,14].

The application of the aerobic treatment, the moving-bed biofilm reactor (MBBR) (intermediate LFL) evaluated by Loukidou and Zouboulis [19], achieved the highest COD removal (81%) compared to all the other aerobic reactors. On the other hand, among physical/chemical treatment, the highest ammonia removal (99.5%) was achieved by air stripping method (old LFL) [26]. For an integrated system of LFL treatment, the application of anoxic/oxic (A/O) combined with membrane bioreactor (MBR) (old LFL) revealed the highest ammonia removal (99.04%) [31] while COD removal (81%) achieved by the combined method of sodium persulfate and hydrogen peroxide (intermediate LFL) [30].

For the system that utilized sequential oxygen supply (sequencing batch reactor), it was proven that COD removal was higher in treating old LFL. However, for the system that fully utilized aerobic treatment (moving-bed biofilm reactor, activated sludge reactor, aerobic lagoon, and rotating biological contactor), the intermediate LFL proved to achieve higher COD removal compared to old LFL as presented in Table 2. Furthermore, for physical/chemical treatment, adsorption treatment was suitable to treat old LFL with COD removal of 69% compared with chemical precipitation (27%) and coagulation/flocculation (10–25%) as shown in Table 3. Nevertheless, for the combined system, the treatment for intermediate LFL (combined sodium persulfate/hydrogen peroxide based advanced oxidation process) achieved slightly higher than old LFL (two-stage anoxic/oxic (A/O) combined membrane bioreactor (MBR)).

Table 1. Properties and waste composition of LFL.

Types of Landfill Leachate	COD (g/L)	BOD_5 (g/L)	BOD_5/COD	pH	Specific Conductivity (µs/cm)	Alkalinity	Heavy Metals (mg/L) Zinc (Zn)	Copper (Cu)	Cadmium (Cd)	Nickel (Ni)	Chromium (Cr)	Age (years)	References
Young	0.41–15	0.036–0.984	0.5–1.0	<6.5	<28,430	<9682	<7.64 [a]	<2.42 [b]	<0.007 [b]	<0.66 [b]	<1.44 [b]	<1	[2,12,13]
Intermediate	0.19–15	0.006–0.98	0.1–0.5	6.5–7.5	2606–41,500	10–2100	<1.43 [b]	<0.39 [b]	<0.03 [b]	<0.37 [b]	<0.28 [b]	1–5	
Old	0.70–10.4	1.5–3.0	<0.1	>7.5	<15,030	1754–5573	<0.003 [b]	<0.15 [b]	<0.04 [b]	<1.34 [b]	<0.002 [b]	>5	

[a]: Exceed the effluent discharge range EPA standard; [b]: fall within the effluent discharge range EPA standard.

Table 2. Variations in the efficiency of aerobic treatment for LFL.

Type of Reactors	Leachate Type [a]	Operational Variables Temperature (°C)	HRT (days)	Removal Rate (%) Nitrogen	COD	Advantages of Reactor	Scale of Study	References
Sequencing batch reactor (SBR)	Old	40–50	20–40	99	75	Suitable for nitrification and denitrification, simple construction and low capital cost.	Pilot scale	[14–18]
	Old	18–25	2–12	70–82	71.2–76.2		Laboratory scale	
	Intermediate	20 ± 0.5	5.63–5.8	90	30–40		Laboratory scale	
Moving-bed biofilm reactor (MBBR)	Old	21	1	-	75	No long settling times for sludge, and less sensitivity to toxic compounds.	Laboratory scale	[14,17,19]
	Intermediate	-	20–24	-	81		Laboratory scale	
Activated sludge reactor (ASR)	Old	21	6.25	-	46–64	Low processing costs and can effectively eliminate biodegradable organic matter by converting it to carbon dioxide and water.	Laboratory scale	[14,20]
	Intermediate	24	0.42	-	75		Laboratory scale	
Aerobic lagoon (AL)	Intermediate	13.5	56	-	75	Low operation and maintenance costs.	Pilot scale	[17,21]
Rotating biological contactor (RBC)	Old	-	1	-	52	Easy to operate, short start-up, minimal land area, low energy consumption, low operating and maintenance costs.	Laboratory scale	[22,23]

[a]: Leachate type is characterized based on pH value.

Table 3. Variations in the efficiency of physical/chemical treatment for LFL.

Adsorption				
Leachate Type [a]	**Adsorbent**	**COD Removal (%)**		**References**
Intermediate	Powdered activated carbon	38		[14]
Old	Peat soil	69		[24]
Chemical precipitation				
Leachate Type [a]	**Precipitant**	**COD removal (%)**		**References**
Old	$Ca(OH)_2$ (1 g/L)	27		[14]
Young	Struvite ($Mg:NH_4:PO_4$ = 1:1:1)	50		[25]
Coagulation/Flocculation				
Leachate Type [a]	**Coagulant**	**Concentration Range (g/L)**	**COD Removal (%)**	**References**
Old	$Al_2(SO_4)_3$	0.7	10–25	[26]
Intermediate	$FeCl_3 + Al_2(SO_4)_3$	1.0–5.0	75	[27]
Air Stripping				
Leachate Type [a]	**Time Stripping (days)**	**Temperature (°C)**	**Ammonia Removal (%)**	**References**
Old	5	-	99.5	[26]
Old	1	20	89	[28]

[a]: Leachate type is characterized based on pH value.

Table 4. Variations in the efficiency of combine treatment for LFL.

Example of Treatment	Leachate Type [a]	Operational Variables	Removal Efficiency	Advantages of Reactor	Critical Remarks/Scale of Study	References
Aerobic Sequencing Batch Reactor (ASBR) combined with zeolite technology	Old	• Temperature (°C): Room temperature • HRT (days): 1 • % Zeolite: 10%	Ammoniacal nitrogen: • ASBR: 65% • ASBR + Zeolite adsorption: 96% COD: • ASBR: 30% • ASBR + Zeolite adsorption: 43%	• Obtained outstanding performance for improved ammoniacal nitrogen removal. • It is also able to eliminate heavy metals and other contaminants that exist in leachate.	• Zeolite is a good adsorbent for treating the ammoniacal nitrogen and COD in leachate. • Laboratory scale.	[29]

Table 4. *Cont.*

Example of Treatment	Leachate Type [a]	Operational Variables	Removal Efficiency	Advantages of Reactor	Critical Remarks/Scale of Study	References
Combined sodium persulfate/Hydrogen peroxide based advanced oxidation process	Intermediate	• Sodium persulfate/hydrogen peroxide ratio (g/g): 1/1.47 • Sodium persulfate dosage (g/mL): 5.88/50 • Hydrogen peroxide dosages (g/g): 8.63/50 • Reaction time (mins): 120	Hydrogen peroxide alone: • Ammoniacal nitrogen: 28% • COD: 31% Sodium persulfate alone: • Ammoniacal nitrogen: 46% • COD: 45% Sodium persulfate followed by hydrogen peroxide: • Ammoniacal nitrogen: 50% • COD: 62% Hydrogen peroxide followed by sodium persulfate: • Ammoniacal nitrogen: 42% • COD: 55% Sodium persulfate combined with hydrogen peroxide: • Ammoniacal nitrogen: 83% • COD: 81%	• Achieved great removal efficiencies for COD and ammoniacal nitrogen than the other processes that used a single oxidizing agent. • Perform more efficiently than the sequential use of sodium persulfate accompanied by hydrogen peroxide in advanced oxidation methods.	• Although persulfate reagent can function independently as an oxidant, its efficiency for oxidizing old leachate is restricted. • Laboratory scale.	[30]
Two-stage anoxic/oxic (A/O) combined membrane bioreactor (MBR)	Old	• HRT (days): 7 • Sludge reflux ratio: 100% • Mixed liquid reflux ratio: 150%	• Ammonia: 99.04% • Total nitrogen: 74.87% • COD: 80.60%	• The results of the mass balance analysis revealed that the second process A/O was absolutely essential in the reduction of the pollutants. • *Pseudomonas*, *Nitrosomonas*, *Planctomyces*, *Nitrobacter*, and *Saprospiraceae* were the most representative genus for denitrification and ensuring nitrogen removal.	• Acclimatization of the activated sludge was carried out by gradually increasing the system's loading in the beginning because the system might not be able to handle the high pollutant loading that was caused by the high concentration of LFL. • Laboratory scale.	[31]

[a]: Leachate type is characterized based on pH value.

Additionally, according to Ahmad et al. [12], membrane procedures are also part of the leachate treatments. However, the biological method, especially the anaerobic process, is the primary focus of this review. It has gained a lot of awareness among researchers and engineers mainly because of its ease of operation, higher removal performance of organics, low risk of odors, and renewable energy production potential in the form of methane.

Most of the previous research on LFL treatment has been focused on the investigation of biological treatment of leachate, including aerobic treatment [12–14]. To the best of our knowledge, no recent review has highlighted the treatment of LFL through the extended use of an integrated AD system, pre-treatment methods, and additive substances. Aiming at the continuous assessment and further evaluation on AD of LFL in previous research works, this review also addressed inhibition factors, potential energy generation as well as kinetics model, and machine learning application. This article provides the latest application and transformation on the treatment of LFL using AD system, serving as a precursor for commercial scale-up and efficient approaches.

In the process of AD, bacteria work in the absence of oxygen to decompose organic matter, as reported by Kurniawan et al. [9], which is abundant in animal manure, biosolids from wastewater treatment, and food waste (FW). In recent decades, there has been increasing demand on anaerobic technology since it effectively eliminates pollutants from wastewater, generates energy in the form of methane, and produces a low volume of sludge compared to aerobic processes [12,14]. Marzuki et al. [32] observed that 12.73 kWh/m^3 of energy can be obtained through the anaerobic treatment of chicken slaughterhouse wastewater. Furthermore, according to Jaman et al. [33], the energy generation for the anaerobic treatment of FW, chicken dung, and co-digestion of FW with chicken dung is 122.96 kWh, 126.10 kWh, and 171.13 kWh, respectively. In addition, complex waste that comes from industrial processes and contains harmful compounds can be effectively treated with anaerobic treatment [34]. Anaerobic reactors can be designed and built in a wide variety of forms and sizes, depending on the site and the feedstock conditions. During the collaborative operation of various types of anaerobic bacteria in the reactor, biogas is produced, which is mainly composed of methane (CH_4), (50–75%) and, along with hydrogen sulphide (H_2S), carbon dioxide (CO_2), water vapour, and small amounts of other types of gases. The methane content of biogas could be used for supplying heat, producing electricity, and running cooling systems. Anaerobic treatment of leachate from landfills has the potential to become a practical solution as it requires less space, uses less energy since it does not have to be aerated, thus creating little or no sludge, and promotes methane formation and recovery [35]. Bhatt and Tao [36] investigated various microorganisms involved in the anaerobic degradation step for the organic material in a sanitary landfill, as shown in Figure 3.

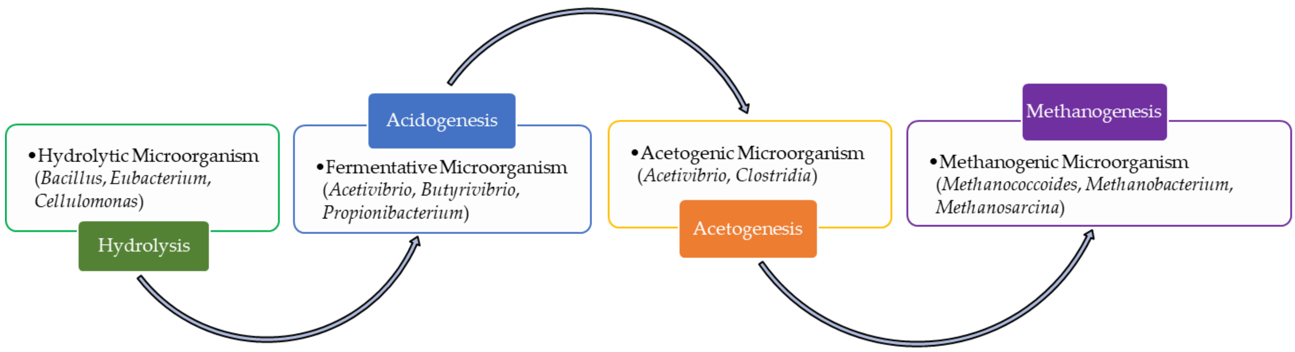

Figure 3. Degradation steps in anaerobic process.

2. Anaerobic Co-Digestion of Leachate with Industrial Wastewater and Solid Waste

2.1. Anaerobic Co-Digestion of Leachate with Industrial Wastewater

2.1.1. Anaerobic Co-Digestion of Landfill Leachate and Crude Glycerol

Biological treatment is typically used in treating LFL [9]. However, due to high levels of toxic compounds in LFL, the ability of biological treatment to successfully eliminate refractory substances is restricted [36]. Therefore, there is a need to implement co-digestion in order to enhance the degradation of organic compounds in LFL, as presented in Table 5. A recent study observed that anaerobic treatment of LFL as a single substrate has caused the disruption of overall treatment efficiency due to changes in the properties of the LFL and the existence of inorganic salts, dissolved organic substances, and heavy metals [37]. Takeda et al. [38] reported that treating wastewater and generating methane is more efficient when combined with substrates that have characteristics that complement one another, as presented in Table 6.

The residual glycerol created by the transesterification process for the production of biodiesel is low in quality and purity. It contains contaminants such as free fatty acids, alcohol, water, organic compounds, catalysts, and soap residue. As a result, its commercial raw material market is therefore constrained. Glycerol waste from the biodiesel process is usually disposed of in landfills or wastewater due to its low purity for industrial purposes [39]. Typically, 10 lb of crude glycerol is produced for every 100 lb of biodiesel produced. A surplus of crude glycerol is produced due to the quick expansion of the biodiesel industry. Thus, biodiesel producers must look for cheaper ways to dispose of this glycerol because it is expensive to purify for use in the food, pharmaceutical, or cosmetic industries [40,41].

Leachate allows great reactor stability since it provides an alkalinity supplement to the treatment system. It is also able to minimise the accumulation of volatile fatty acids (VFA) and is capable of diluting the toxic compounds of glycerol (methanol), since it contains a high level of moisture content [38,42]. Moreover, it acts as a supply of macronutrients and micronutrients required for microbial growth, while also offering a greater balance of carbon to nitrogen (C/N) ratio [38,42–44]. On the other hand, glycerol offers a high amount of readily biodegradable organic material and improves the C/N ratio during the anaerobic treatment of LFL [37,38,44]. Therefore, it can be concluded that the addition of glycerol as a co-substrate into the AD of LFL will balance the nutrient content and enhance the amount of biogas.

Several volumes of LFL and glycerol were investigated to optimize organic material reduction and methane production. Takeda et al. [38] reported that a Central Composite Rotational Design (CCRD) was carried out and obtained the highest COD removal efficiency of 96.46%, with the lowest level of substrate/inoculum (S/I) ratio (0.23 gCOD/g VSS), time (25 days), and glycerol content (1.1%). From the study conducted, the cumulative specific biogas production ranged between 104.21–312.37 mL/g VSS. The highest specific biogas production was 312.37 mL/g VSS, with a S/I ratio of 0.5 gCOD/g VSS and a glycerol content of 1.5%. Hence, it shows that adding the glycerol (optimum glycerol content = 1.5% with an optimum S/I ratio = 0.50 gCOD/g VSS) to the AD of LFL increases the removal of organic material and generation of biogas compared to the mono-digestion of LFL.

Table 5. Performance of co-digestion.

Types of Anaerobic Digestion	HRT (Hours)	Volume of Leachate	COD Removal Efficiency (%)	BOD/COD	Specific Biogas Production (mL/g VSS)	Specific Biomethane Production (mL CH_4/g VSS)	Methane Production	Reference
Anaerobic Co-Digestion of Leachate with Industrial Wastewater								
Anaerobic co-digestion leachate and crude glycerol (crude glycerol content: 1.50%)	720	-	92.03 (Soluble COD)		312.37	244.59	78.3%	[38]
Anaerobic co-digestion of landfill leachate and acid mine drainage	20	-	83		-	-	1805 (mL/d)	[45]
Anaerobic co-digestion of leachate with solid waste								
Anaerobic co-digestion of food waste and landfill leachate	840	568 mL	-	1.48	878	-	466 mL/g VS	[46]
Anaerobic co-digestion of sewage sludge with landfill leachate	319.2	100 (mL/d)	-	1.07	-	-	375 L	[47]

Table 6. Properties of substrate and co-substrate.

Leachate	Crude Glycerol	References
• Low concentration level of phosphorus	• High concentration level of phosphorus	[38]
• High moisture content	• Low moisture content (toxic compound)	[38,42]
• High content of macro and micronutrients	• Low content of macro and micronutrients	[38,42,43]
• Contains high recalcitrant substances/high amount of non-biodegradable matters	• High biodegradable organic load/serves readily biodegradable organic material	[37,38,44]
• High ammoniacal nitrogen content	• Low nitrogen content	[38,43,44]
Leachate	**Acid mine drainage**	**References**
• pH modifier agent • High carbon content	• Low pH • High sulphate content	[45]
Leachate	**Food waste**	**References**
• High water content • Low biodegradability • pH modifier agent	• Low water content • High biodegradability • Low pH	[46]
Leachate	**Sewage sludge**	**References**
Optimum mixing ratio: 20:80		[47]

2.1.2. Anaerobic Co-Digestion of Landfill Leachate with Acid Mine Drainage

Zhou et al. [45] found that both acid mine drainage (AMD) and LFL have properties that can improve the nutritional balance in the anaerobic process. AMD is a source of sulphates and according to Zan & Hao [48], regulating sulphates in the anaerobic co-digestion process can be one method to improve the production of methane.

AMD is formed from mining activity and is normally associated with coal mining, which contains highly acidic water and is rich in heavy metals. AMD has a low level of pH, contains saturated heavy metals, and is high in sulphate contents [45,49,50]. Addition of sulphate into LFL treatment can improve the biodegradation of propionic acid and the generation of methane [45,51,52]. Moreover, the addition of sulphate can easily break down the biodegradable substrates by 93% [48]. On the other hand, LFL has the potential to be applied in the treatment of AMD as it provides a source of carbon. However, in this review, only LFL treatment is discussed.

Anaerobic co-digestion (ACoD) of LFL and AMD was performed in up-flow anaerobic sludge blanket reactor (UASB) [45]. Researchers investigated effect of different hydraulic retention times (HRT) (30 h, 20 h, 12 h, and 8 h) were evaluated. From the results obtained, HRT of 30 h, the removal of COD was only 78% with the methane production of 1700 mL/d. When HRT dropped to 20 h, methane production increased to 1805 mL/d, and COD removal increased to 83%. However, when HRT decreased to 10 h, removal efficiency for COD decreased to 71%, and methane production was 1589 mL/day. This reveals that HRT plays a significant role in the AD since it has a certain effect on the removal of COD and the generation of methane. Hence, the optimum HRT in order to achieve a higher efficiency in organic removal and methane production is 20 h.

2.2. Anaerobic Co-Digestion of Leachate with Solid Waste

2.2.1. Anaerobic Co-Digestion of Food Waste and Landfill Leachate

In order to enhance the AD of FW, co-digestion of FW with LFL can be done and compared in terms of biogas and methane production. FW provides an opportunity for the generation of biogas. However, accumulated VFA frequently make it possible to limit the generation of methane due to their extremely high biodegradability. As an alternative, LFL was employed as a co-substrate to improve the effectiveness of the anaerobic processes of FW [46].

From the experiment conducted by Liao et al. [46], the co-digestion of FW and LFL was done in single-stage batch reactors with a working volume of 1500 mL for 35 days (HRT). The reactors were kept at a temperature of 35 $\pm$ 1 °C (mesophilic condition) in a water bath. There are eight reactors fed with the same amount of FW but different volumes of leachate. The most biogas and methane were achieved with 568 mL of leachate added as a co-substrate. The least biogas and methane were achieved with a leachate content of 142 mL. The ratio of BOD/COD (1.48) of co-digestion of FW with LFL indicates that the sample has high biodegradability [46], as shown in Table 5.

The co-digestion of FW with LFL was also conducted by Dearman and Bentham [53], Shahriari et al. [54], and Stabnikova et al. [55], which employed leachate recirculation to enhance the AD of FW.

2.2.2. Anaerobic Co-Digestion of Sewage Sludge and Landfill Leachate

The ACoD of LFL and sewage sludge (SS) was noticed to be very practicable for the production of methane [47]. The addition of leachate to mesophilic AD produces more methane than mono-digestion of SS. There are 2 phases conducted, which consist of different amounts of LFL added to the AD system. In phase 1, the amount of leachate was under 12% of the SS volume, while in phase 2, the amount of leachate was under 25% of the SS volume. The addition of leachate as a co-substrate resulted in higher methane production (methane volume = 350 L) in phase 2 with a leachate volume of 100 mL/d, whereas in phase 1 with a leachate volume of 60 mL/d, the methane production was lower (methane volume = 115 L). Nonetheless, during phase 2, mono-digestion of SS produced the most methane (methane volume = 399 L) compared to co-digestion. It shows that at higher volumes of leachate, the methane production was not higher than in the control reactor (mono-digestion). This could be due to shorter retention times and a reduce in total volatile solids removal. In addition, it was observed that adding leachate to the SS would not increase the concentration of heavy metals in the sludge biosolids. From the result obtained, the optimum volume of leachate to be added to SS is 100 mL/d, with a production of methane of 375 L. Therefore, the ACoD of LFL and SS is a promising alternative to enhance the production of methane.

Berenjkar et al. [56] and Montusiewicz and Lebiocka [57] also investigated the co-digestion of LFL with SS to evaluate the maximum biogas yield that can be generated.

3. Pre-Treatment of Landfill Leachate

LFL contains various different contaminants and is rich in suspended solids, organic and inorganic compounds, and heavy metals. Discharge of improperly treated LFL might be a major cause of water pollution and emissions of polluted gas in air [58]. Conventional approaches, such as biological treatment, are insufficient to treat the polluted LFL and are also unable to eliminate the adverse environmental impact [14]. Reported studies have indicated that pre-treatment technologies are effective in removing suspended solids, break down organic matter and ammoniacal nitrogen, minimise toxicity, and enhance the biodegradability of the LFL [20].

3.1. Coagulation/Fenton/Air Stripping

Smaoui et al. [6] and Guo et al. [10] conducted a research on several processes for the preliminary treatment of LFL in order to improve anaerobic treatability of LFL. Researchers compared the pre-treatment performance of coagulation-flocculation, Fenton oxidation (FO), and air stripping. As shown given in Figure 4, the BOD_5/COD ratio improved from 0.28 to 0.32 (air stripping), from 0.28 to 0.37 (CF), and from 0.28 to 0.39 (FO) [6,10]. As a result, it can be concluded that both FO and CF displayed great potential in terms of removing organic matter and improving the biodegradability of the effluent.

Batch AD was done to examine further on how each pre-treatment affects the production of biogas. The biogas production was monitored for 50 days and is done in a mesophilic environment (37 ± 1 °C). Smaoui et al. [6] performed five sets of batch mode experiments: (1) anaerobic sludge without LFL; (2) anaerobic sludge with raw LFL; (3) anaerobic sludge with LFL treated by CF; (4) anaerobic sludge with LFL treated by FO; and (5) anaerobic sludge with LFL treated by air stripping were observed. Table 7 shows that the highest amount of methane was formed by air stripping (588 mL/g COD_{in}), followed by FO (448 mL/g COD_{in}) and CF (370.9 mL/g COD_{in}). The raw leachate resulted in the least amount of biogas generation (163.69 mL/g COD_{in}) among the other result. Therefore, it can be said that extensive pre-treatment is necessary to increase production of methane. Thus, it can be concluded that air stripping was the best pre-treatment method because it produced the most methane compared to CF and FO pre-treatments.

3.2. Electrochemical Oxidation

Pasalari et al. [59] and Fernandes et al. [60] stated that electrochemical oxidation (EO) is one of the pre-treatment processes for LFL. They stated that, EO helps to improve biodegradation and biogas. LFL has organic waste that can be turned into energy with the help of pre-treatment technologies and biological treatments [58]. EO is a promising technique that increases biodegradability since it can transform high-molecular compounds into low-molecular, and is simple to conduct and operate. Therefore, it is conceivable to consider this technology as a pre-treatment step with the goal of increasing methane production during the ACoD process [60].

In a batch mode reactor, electrochemical oxidation tests were conducted to improve the biodegradability of raw LFL before it was fed into ACoD (LFL with sludge). The tests were run at a temperature of 20 °C. Since the EO process was considered as a pre-treatment method, a low current density was adopted in order to achieve the electrochemical conversion of recalcitrant organic matter in raw LFL, such as humic acid. The experimental batch tests were performed to find out the biochemical methane potential (BMP) of co-digested substrates with different ratios (15%, 25%, and 35%) of raw LFL in both reactors (treated with EO and controls reactors).

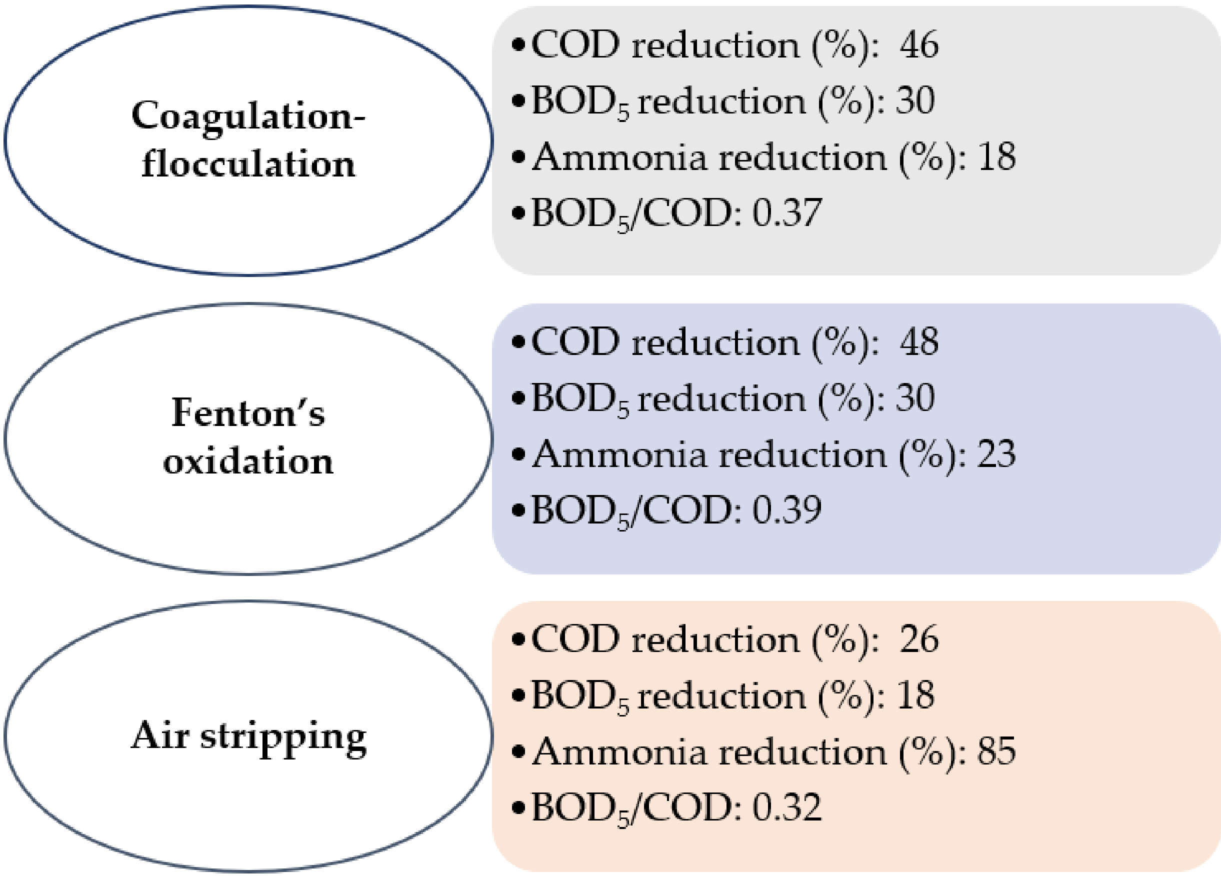

Figure 4. Reductions in COD, BOD_5, and NH_3 attained in each pre-treatment stage prior anaerobic digestion.

From the results observed in Table 7, the methane yield in ACoD reactors pre-treated with EO is higher than the control. The highest methane was produced from R6 (0.2925 L/g $sCOD_{removed}$), while the least was produced by R1 (0.1020 L/g $sCOD_{removed}$). The methane yields in ACoD reactors pre-treated with EO showed increasing trends in the range of 0.1368 to 0.2925 NL/g $sCOD_{removed}$ as the volume of influent raw LFL increased (150 mL, 250 mL, and 350 mL). As a result, ACoD of LFL that has been treated with EO and sludge might be proposed as a technology, as it has a lot of potential and is relatively inexpensive [60,61].

3.3. Coagulation-Adsorption

Physical and chemical processes are the most efficient methods for pre-treating the LFL [62]. In order to achieve high COD removal, a combination of pre-treatments was conducted. The coagulation process was conducted in young and old leachate, while the adsorption process was used for only old leachate. In the coagulation process, alum and ferric chloride ($FeCl_3$) were used, while fly ash was used in the adsorption process.

In treating the old leachate by using the coagulation process, it was observed that the COD removal efficiency increased with the increase in doses of $FeCl_3$ and alum. The dose of $FeCl_3$ increases from 0.2 to 0.7 g/L and afterwards remains constant for COD removal, while the dose of alum increases from 0.2 to 0.6 g/L. Nonetheless, when the dose is more than 0.6 g/L, it caused COD removal to slightly drop. The highest COD removal by using $FeCl_3$ and alum was 59% and 75%, respectively, as presented in Table 7. In addition, when treating young leachate, it was found that the elimination of COD rises from 0.2 to 0.6 g/L for $FeCl_3$ and from 0.2 to 0.8 g/L for alum and remains unchanged after that. The highest COD removal by using $FeCl_3$ and alum was 35% and 55%, respectively. During the adsorption process, the COD removal efficiency of old leachate is found to be 28%, and the optimum amount of fly ash to use is 6 g/L.

Therefore, among the pre-treatment methods, the highest COD removal rate for pre-treatment of LFL in an air stripping system is 85%, compared with coagulation (FeCl = 59%, and alum = 75%) and adsorption process is 28% as shown in Table 7.

Table 7. Performance on anaerobic digestion of landfill leachate after pre-treatment processes.

Type of Pre-Treatment	**Coagulation/Fenton/Air Stripping**				**Remarks/Scale of Study**	**References**
	Parameter					
	pH	**COD removal (%)**	**Biogas yield (mL/g CODin)**			
Coagulation-flocculation	7.96	75	370.90		• COD of sample: 40,000–44,000 mg/L • Pilot scale	[6]
Fenton's oxidation	8.09	77	448.00			
Air stripping	8.29	85	588.88			
Raw LFL	7.93	68	163.69			
Type of pre-treatment	**Electrochemical Oxidation (EO)**					
	Parameter					
	Leachate (mL)	**Inoculum (mL)**	**Methane yields (NL/g sCOD removed)**	**Methane content (%)**		
Control:					• COD of sample: 320–1165 mg/L • Laboratory scale	[59]
System 1	350	200	0.1712	48		
Assisted with EO pre-treatment:						
System 2	350	200	0.2925	54		
Type of pre-treatment	**Coagulation and Adsorption**					
	Parameter					
Coagulation:					• COD of sample: 6240–66,240 mg/L (Young LFL) and 1024–19,200 mg/L (Old LFL) • Laboratory scale	[62]
Type of leachate	**Ferric chloride dosage (g/L)**	**COD removal (%)**	**Alum dosage (g/L)**	**COD removal (%)**		
Old leachate	0.7	59%	0.6	75%		
Young leachate	0.6	35%	0.8	55%		
Adsorption:						
Type of leachate	**Fly ash dosage (g/L)**		**COD removal (%)**			
Old leachate	6		28%			

4. Anaerobic Digestion of Landfill Leachate

4.1. Anaerobic Reactor

Various types of anaerobic reactors have been extensively studied for the removal of pollutants from LFL and generation of energy in the form of methane. Completely mixed anaerobic digesters, UASB, anaerobic filters (AF), and fluidized and expanded bed reactors are the most common types of anaerobic reactors [61]. According to Ahmad et al. [12], anaerobic membrane bioreactors (AnMBR) and anaerobic contact reactors are also anaerobic reactors used to treat the LFL. Among the important operating factors that need to be considered for the design and operation of AD reactors are leachate type, COD content, HRT, and organic loading rate (OLR), as shown in Table 8. Anaerobic fluidised bed reactors (AFBR) and AnMBR achieved higher COD removal (90%) compared to other anaerobic reactors. The methane formation produced by AFBR was 75% [63]. According to Zayen et al. [64], AF generated 19.24 L/d of methane.

4.2. Design Parameters in LFL Anaerobic Treatment

The effectiveness operation of AD systems is affected by several design parameters, as illustrated in Figure 5. Among them, pH is a significant parameter in AD since the microbial activities are extremely sensitive to changes in pH levels. According to Nain et al. [68], the optimal pH for AD of LFL should be in a range of 5.56–7.58. Thus, precise control of pH levels in LFL treating anaerobic digesters is a top requirement [2]. Ahmad et al. [12] found that sodium hydroxide and sodium bicarbonate can be used to regulate pH in anaerobic treatment systems. The ideal temperature for the growth of bacteria in anaerobic treatment is typically between 25 to 35 °C. If the temperatures are below the ideal range, the removal efficiency will decrease.

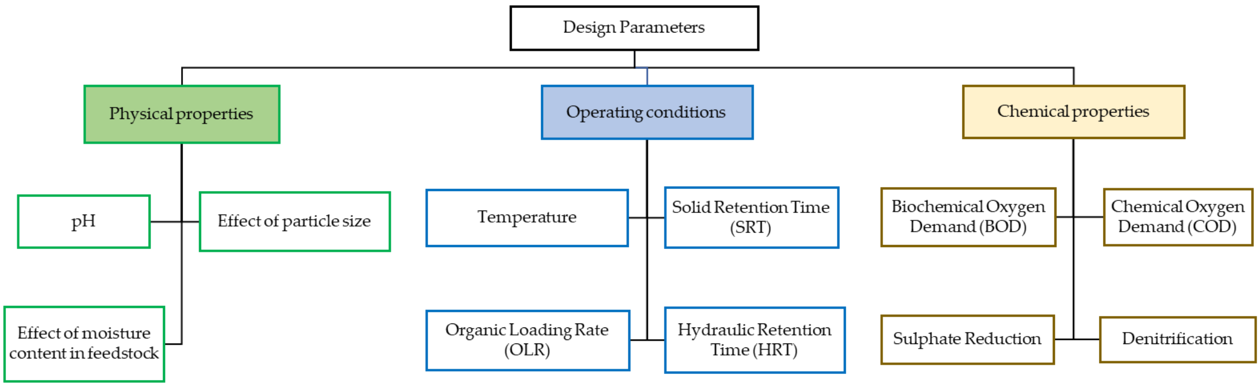

Figure 5. Design parameters in anaerobic digestion system.

Lower HRT and greater OLR are desirable for treating low concentrations of wastewater to ensure that the microbes have access to nutrients. Lower OLR is recommended for treating wastewater with high concentrations in order to complete biodegradation of the substrate and avoid sludge flotation [69]. The optimum OLR is 9.6 kg COD/m^3.d with COD removal is 90% [69]. In addition, BOD concentration is another factor affecting the performance of AD. It can show how much biodegradable organic matter there is, which is an important aspect to consider during anaerobic treatment [70].

In addition, the optimal moisture content in feedstock is also a significant factor in the AD system since it affects methane yield. Lohani and Havukainen [71] stated that for a better kinetic process and methane yield, the particle size should be small and the solid retention time (SRT) must be long enough to ensure an adequate level of methanogenic activity. Furthermore, the effect of sulphate reduction on AD systems is a crucial factor that needs to be taken into account since it can inhibit nearly all microbial groups. Lastly, the presence of nitrate in AD needs to be looked into, as it has a significant effect on microbial competition, which decelerates methane production.

5. Integrated System for Anaerobic Treatment of Landfill Leachate

There are several studies showing by integrating chemical, physical, and biological processes in any order can improve the efficiency of LFL treatment. From the review conducted by Ahmad et al. [12], there is no specific technology claimed to be adequate for the whole treatment; hence the necessity of implementing the integrated system in LFL treatment. Table 9 depicts the efficiency in adopting the integrated systems in LFL treatment that have been discovered from previous studies.

Un et al. [72] conducted a study to compare the efficiencies of anaerobic batch reactor (ABR) alone and integrated with electrocoagulation (EC) for the treatment of LFL. From the study, it was found that 74% of COD was removed by single step anaerobic treatment while integration of EC prior to ABR enhanced COD treatment efficiency by 92%. It can be seen that by using EC as a pre-treatment of LFL is the most appropriate and desirable technique to enhance the AD process of LFL. This is primarily due to the fact that EC has the potential to remove non-biodegradable COD, requires less coagulant, less sludge production, and easy to operate using simple equipment.

Table 8. Performance of different type of anaerobic reactors.

Type of Anaerobic Reactors	Characteristics						Critical Remarks/Scale of Study	COD Concentration in Effluent (mg/L)	References
	Leachate Type	Chemical Oxygen Demand, COD Content (mg/L)	Hydraulic Retention Time, HRT (days)	Organic Loading Rate, OLR	Removal Efficiency	Methane Production			
Upflow anaerobic sludge blanket reactors (UASB)	Old	14,640	30	-	• COD: 74% • TP: 89% • TSS: 81% • BOD: 64% • TN: 50%	-	• Has great ability in removing TP and TSS in leachate. • There is no need for support material and concentrated biological growth. • Not really efficient for TN removal. • Creates low sludge output. • Laboratory scale. • Volume of sample treated: 40 cm	3806.4 > 250 [a]	[65,66]
Anaerobic fluidised bed reactors (AFBR)	Young	35,000 (avg)	1	12 g COD/L/day	• COD: 90%	75%	• Can accumulate a substantial quantity of biomass by natural attachment. • Short retention times. • High flow rates. • Has great stability performance. • Pilot scale. • Volume of sample treated: 30 and 75 cm	3500 > 250 [a]	[63]

Table 8. *Cont.*

Type of Anaerobic Reactors	Characteristics						Critical Remarks/Scale of Study	COD Concentration in Effluent (mg/L)	References
	Leachate Type	Chemical Oxygen Demand, COD Content (mg/L)	Hydraulic Retention Time, HRT (days)	Organic Loading Rate, OLR	Removal Efficiency	Methane Production			
Anaerobic Filter (AF)	Young	15,200	4.5	3.3 g COD/L/day	• COD: 74.72%	19.24 L/d	• High-load systems, stable under transient condition (fluctuations in effluent compound and toxic substances are present) and shows a great performance in terms of COD removal and the generation of biogas. • Pilot scale. • Working volume of reactor: 20 L	3842.56 > 250 [a]	[64]
Anaerobic membrane bioreactors (AnMBR)	Old	39,000 (avg)	2	2.5 kg COD/m^{3}d	• COD: 90%	-	• MBR system was run with a mix of leachate and synthetic wastewater. • Submerged membrane reactors provided more compact systems and save energy. • Less sludge production. • RO process and stripping have been used for post-treatment since the quality of MBR effluent is poor. • Laboratory scale. • Working volume of reactor: 29 L	3900 > 250 [a]	[67]

[a]: Effluent discharge standard based on US EPA.

Fazzino et al. [73] investigated the combination processes of active filtration and AD to treat mature LFL. Researchers applied active filtration using zero-valent iron (ZVI) mixed with lapillus and ZVI mixed with granular activated carbon (GAC) to remove the heavy metals. The removal efficiencies of COD, Cu, Ni, and Zn obtained from the ZVI/lapillus filter were 33%, 85%, 66%, and 58%, respectively, while treatment efficiencies increased to 56%, 91%, 67%, and 75% for COD, Cu, Ni, and Zn, respectively using ZVI/GAC filter. Thus, these results indicate that ZVI/GAC has a better performance in pre-treatment of LFL.

Wang et al. [74] performed the treatment of LFL by implementing anoxic/aerobic granular active carbon assisted membrane bioreactors (A/O-GAC-MBR) integrated with nanofiltration (NF) and reverse osmosis (RO). The presence of GAC enhances the reduction of harmful organic pollutants and heavy metals. Additionally, it increases bio flocculation and flocs' size, which considerably reduce membrane fouling. Moreover, the application of NF and RO membranes were utilized as further treatment of MBR effluents, where the NF being most effective in removing colour with 93.75% of removal.

Ozone direct oxidation pre-treatment and catalytic oxidation post-treatment coupled with an anaerobic baffled membrane bioreactor (ABMBR) in treating LFL, was conducted by Yuan et al. [75]. From the integrated treatments method, the total reduction of COD and ammonia nitrogen were 91.2% and 99.4%, respectively which was higher than removal efficiencies of ABMBR treatment; 80.38% and 21.56%, respectively.

Li et al. [76] applied combined process for the treatment of mature LFL in a full-scale treatment system. In the combined process, including the sequencing batch reactor (SBR), was used as primary treatment, followed by polyferric sulphate (PFS) coagulation and the Fenton system for secondary treatment, and a pair of up-flow biological aerated filters (UBAFs). After combined treatment, the total reduction of COD and ammonia were 97.3% and 99%, respectively.

The investigation by Bakraouy et al. [77] indicated that the combination of anaerobic treatment with the CF process was effective in treating LFL. The $FeCl_3$ was used as a coagulant, while the cationic polymer was used as a flocculant. The COD elimination efficiency rises linearly with coagulant and flocculant dosages. However, adding reagents at a certain concentration does not improve removal efficiency. The optimum dosages obtained were 4.4 g/L of coagulant and 9.9 mL/L of flocculant, with the total reduction of phenol, turbidity, colour, and COD were 89%, 69%, 94%, and 80%, respectively. Therefore, it shows that the combined process of SBR, PFS coagulation and the Fenton system, and a pair of UBAFs appears a higher COD and ammonia removal (97.3% and 99%, respectively) compared to other integrated systems. For treating old LFL by using integrated system (i.e., combined process including sequencing batch reactor (SBR), with polyferric sulfate (PFS) coagulation and the Fenton system, and a pair of up-flow biological aerated filters (UBAFs)) proven that 17.3% higher of COD removal was achieved compared with young LFL.

Post Treatment for Ammonia Removal in Landfill Leachate

Post treatment of LFL, particularly mature LFL, is required to eliminate excessive concentrations of ammonia and organics in LFL in order to prevent environmental pollution through a process known as anaerobic ammonium oxidation (Anammox) [78]. Annamox is particularly suited for the treatment of nitrogen-rich wastewaters, such as LFL. Anammox bacteria, converts ammonium into dinitrogen gas as depicted in Figure 6 by using ammonium nitrogen as an electron donor and nitrite as an electron acceptor under anoxic conditions [79]. It was demonstrated that treatment based on Anammox is promising for potential application in the elimination of nitrogen from LFL due to its cost-effectiveness (less aeration energy) and the high performance of denitrification [80]. However, according to Jin et al. [81] and Ye et al. [80], the organic substances and the high concentrations of biodegradable organics in LFL have an adverse effect on Anammox bacteria, which inhibits the application of the Anammox process to be broadly used. Furthermore, Anammox bacteria have been shown to be sensitive to the toxicity of a wide range of organic matters, including aromatic compounds (phenols and quinolines) and antibiotics (norfloxacin and enrofloxacin), which may have a negative impact on the massive number of functional genes and proteins involved in nitrogen removal [81]. Additionally, LFL containing excessive organic and nitrogen substances must be pre-treated by AD prior to the Anammox process in order to prevent the inhibition initiated by Anammox bacteria.

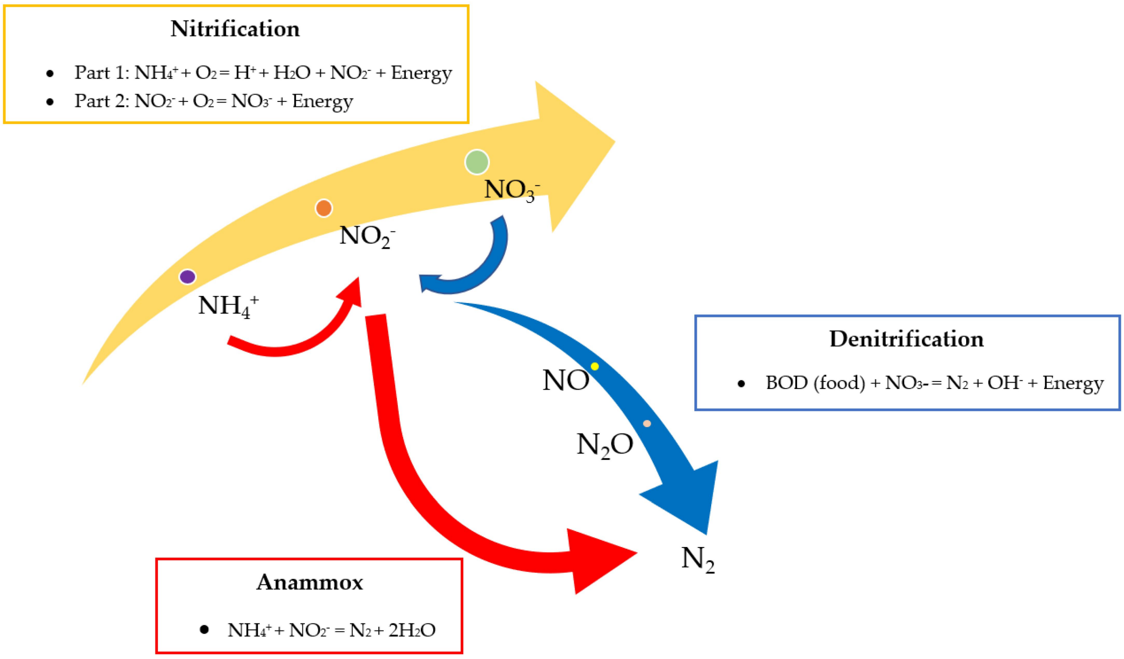

Figure 6. Denitrification using Anammox method and conventional nitrification and denitrification processes.

6. Potential Inhibitors in the Anaerobic Treatment of Landfill Leachate

AD has become widely recognised for treating solid waste and wastewater, with the added benefit of waste-to-energy conversion [82]. There are various affecting factors that need to be taken into account while operating the AD process, such as the carbon to nitrogen ratio and contents of sugar, nitrogen, salinity, carbon, and trace elements of the substrate.

According to Lohani and Havukainen [71], carbon, nitrogen, and phosphorus ratio (C:N:P) could influence the production of methane, and the recommended ratio is 100:3:1 for generating a high methane yield. A substrate with imbalance C/N ratio will reduce the methanogenesis process due to low pH (<6.8) and the accumulation of VFA, which then affect methane production [83–85]. In addition, Jiang et al. [86] reported that a sample with a low concentration of ammonium in a range between 50 and 200 mg/L is favourable for anaerobic processes since the ammoniacal nitrogen is required to synthesis amino acids, proteins, and nucleic acids, while if the sample has higher concentrations of ammonia, the methanogenesis activity in an AD reactor will be inhibited. Náthia-Neves et al. [87] conclude that a substrate with a higher C/N ratio has excess carbon and this causes quick consumption of nitrogen by methanogenesis and lower biogas production.

According to Liu et al. [88], the level of salinity in the sample is an important factor in the microbial activity of the AD process. From the study shows that, a low salinity level promotes the processes of hydrolysis and acidification in AD but inhibits the methanogenesis process. On the other hand, when salinity levels are extremely high, the acidification and methanogenesis processes are severely hindered. For instance, the degradation efficiency of acetate dropped from 53.9% to 12.6% when the level of salinity (NaCl) increased from 0 to 15.0 g/L, as evaluated by Zhao et al. [89]. This demonstrates that the higher content of NaCl causes failure in the AD process and inhibits methane production.

Table 9. Efficiency in adopting the integrated systems in LFL treatment.

Type of Integrated System	Type of Leachate	Pollutant Content	Removal Efficiency	Remarks	References
Integrated Electrocoagulation (EC) [a] and the Anaerobic Treatment [b]	Old	• COD: 6400 mg/L	• COD: 92%	• The result of treating LFL with combine technology between electrocoagulation and AD were shown to be more effective in COD removal than those achieved using each treatment method individually.	[72]
Integrated treatment via Active Filtration [a] and Anaerobic Digestion [b]	Old	• COD: 3500 mg/L • Cu: 2 mg/L • Ni: 2 mg/L • Zn: 5 mg/L	ZVI/lapillus: • COD: 33% • Cu: 85% • Ni: 66% • Zn: 58% ZVI/GAC: • COD: 56% • Cu: 91% • Ni: 67% • Zn: 75%	• ZVI/GAC showed better pre-treatment performance compared with ZVI/lapillus since lapillus was discovered to be an unsuitable material for removing ammonium, chloride, and COD. • GAC worked as an effective organic matter adsorbent, removing organic compounds from LFL but not ammonia nitrogen.	[73]
Anoxic/aerobic granular active carbon assisted MBR [a] integrated with nanofiltration and reverse osmosis [b] (A/O-GAC–MBR integrated with NF and RO membranes)	Old	• COD: 3134.88 mg/L • NH_3-N: 434.76 mg/L	• COD and NH_3-N: >80% (MBRs) • Colour: 93.75% (NF membrane)	• GAC greatly enhance the reduction of heavy metals such as Cd, Cu and Cr and also COD. • NF membrane exhibited remarkable removal efficiency of colour and organic contaminates. • The integrated approach is a viable alternative for large-scale leachate treatment.	[74]

Table 9. *Cont.*

Type of Integrated System	Type of Leachate	Pollutant Content	Removal Efficiency	Remarks	References
Ozone direct oxidation pre-treatment [a] and catalytic oxidation post-treatment coupled with anaerobic baffled membrane bioreactor (ABMBR) [b]	Old	• COD: 12,320 mg/L • NH_4^+-N: 1583.16 mg/L	• COD: 91.2% • NH_3-N: 99.4%	• Ozone direct oxidation was a highly efficient method of pre-treatment compared with potassium peroxymonosulfate (PMS). • The post-treatment of an ABMBR effluent were consist of struvite precipitation, ozone catalytic oxidation and post-MBR process. • The combined treatments of ozone direct oxidation pre-treatment, ABMBR treatment, and series of post treatment is efficient in treating of LFL.	[75]
Combined process including sequencing batch reactor (SBR) [c], with polyferric sulfate (PFS) coagulation and the Fenton system [d], and a pair of up-flow biological aerated filters (UBAFs) [e]	Old	• COD: 3000 mg/L • NH_3-N: 1200 mg/L	• COD: 97.3% • NH_3-N: 99%	• SBR treatment was effective as a primary treatment for the removal of ammonia, biodegradable carbon, and phosphorus. • PFS coagulation and the Fenton system are used for secondary treatments in treating non-biodegradable leachate from the SBR. • Two UBAFs act as a tertiary treatment or final polishing step, which is used as a refining step for the physicochemical treatment. • The combined processes are an effective alternative treatment for small-scale LFL treatment plants.	[76]
Anaerobic digestion [a] combined with coagulation and flocculation (CF) [b] using ferric chloride as coagulant and cationic polymer as flocculant	Young	• Phenol: 341.6 mg/L • Turbidity: 222 NTU • Colour: 0.491 (FD = 20) • COD: 11,520 mg/L	• Phenol: 89% • Turbidity: 69% • Colour: 94% • COD: 80%	• CF has been effectively used as post treatment of AD, and it has many advantages, such as easy to handle and less cost required. • The optimum dosage of coagulant: 4.4 g/L. • The optimum dosage of flocculant: 9.9 mL/L.	[77]

[a]: Pre-treatment of integrated system; [b]: Post treatment of integrated system; [c]: Primary treatment; [d]: Secondary treatment; [e]: Final polishing step.

Lastly, the trace elements are also an important aspect of the AD process. Matheri et al. [90] stated that trace elements, which include Ni, cobalt (Co), calcium (Ca), and potassium (K), are important for microbial growth as they provide the macro and micro nutrients to the microbes in the AD system. Trace elements can be inhibiting, stimulating, or toxic to the AD system, depending on toxic threshold concentration values and their composition in LFL [13,90], presented in Table 10.

Table 10. Recommended threshold value of the trace element to stimulate biogas production and composition of heavy metals in landfill leachate.

Trace Element	Toxic Threshold Concentration (mg/L)	Composition of Heavy Metals in Landfill Leachate (µg/L)	References
Calcium	2800	-	[13,90]
Manganese	50	-	
Copper	400	3–157	
Zinc	1	10–303	
Iron	10	-	
Cadmium	0.18	0.1–35	

7. Application of Additive Substances into Anaerobic System

Use of additive substances such as conductive materials [91] and conductive nanoparticles (CNPs) [92], in AD systems enhances microbial colonization, eliminates toxic compounds, accelerates direct electron transfer (DIET) and promotes methane production as presented in Table 11 [89,93,94].

In addition, the addition of conductive materials maintains the stability of the reactor during high OLR circumstances and is also effective in converting VFA to methane. Bio-based carbon materials, such as carbon cloth, biochar, and activated carbon (AC), are suggested additives for AD systems since they are inexpensive and can be produced directly from biomass. In addition, graphene and carbon nanotubes were also discovered to improve the DIET; however, their production costs are very expensive, making widespread implementation economically impractical. Moreover, iron-based conductive materials such as magnetite can be used in an AD system to help adsorb and eliminate toxic compounds from LFL, which can improve the overall AD system [91].

CNPs are nano-sized structures (1–100 nm) and they have ability to increase AD by reacting with the substrate and microorganisms. The most important factors for CNPs to be used as additives are their physicochemical properties which includes high activity, high reactive surface area, and expedite the hydrolysis or acidification process, thus improve biogas generation [92]. A large specific surface area is a significant factor for conductive materials, as it serves an abundance of attachment sites for the microbial community (or know as microbial iteration) [95]. There are various categories of CNPs employed in the AD system, such as, metal oxides (copper(II) oxide, CuO and zinc oxide, ZnO), zero-valent metals (ZVMs) (Ni) and carbon-based conductive nanoparticles. ZVMs provides hydrogenotrophic methanogens' activities more efficient; thus, by implementing them in the process of converting biomass into methane is beneficial. This activity will speed up the hydrolysis stage. In addition, the metal oxides have shown remarkable success in converting substrate into biogas throughout the AD system [92]. According to Purnomo et al. [96], the large surface area of carbon-based CNPs can encourage chemical reactivity and provide thermal stability in the AD system.

Furthermore, the impact of silver nanoparticles or nanosilver (AgNPs) and engineered nanomaterials (ENMs) on anaerobic systems is also being investigated. AgNPs are generated during industrial processes or as consumer by-products that might be disposed of in sanitary landfills either directly or indirectly [97], while ENMs are frequently found in commercial products and eventually increase the nanoparticles (NPs) in the landfill [98]. Yang et al. [97] reported that the implication of AgNPs for the anaerobic process was observed when the accumulation of AgNPs was higher than 10 mg/kg in landfills. This is because the accumulation of VFA and low pH lead to inhibition conditions, which affected the methanogenic population and the process of generating biogas. On the other hand, Demirel [98] investigated the ENMs such as ZnO with 34.5 mg/L dosage caused the reactor to become unstable and led to a reduction in methane formation. Therefore, the AgNPs and ENMs can cause toxicity to the AD process and disrupt biogas production.

8. Kinetic and Machine Learning Evaluation

Kinetic studies and machine learning assist in understanding how reactions operate, and they are useful to measure AD, estimate the rates of COD removal, evaluate the production of methane, and predict energy generation in a larger scale. According to Jaman et al. [110], kinetic studies are simple to work with and serve as an effective instrument for comparing the predicted data with the experimental results. The most common kinetic models that have been successfully employed for effective optimisation of biological process parameters are the Stover–Kincannon model, first-order kinetics, Van der Meer and Heertjes model, and the Gompertz model, as shown in Table 12 below. However, kinetic studies have limitations; they cannot provide as much information as an artificial intelligence-based (AI) model [111].

Models based on artificial intelligence, such as fuzzy models and artificial neural networks (ANN), are capable of taking into consideration numerous factors that affect the production of methane in the system that is being investigated. The examples of parameters that can be considered by an artificial base model are input and target variables, which are limitations in the Gompertz model [112]. Jaroenpoj et al. [113] stated that artificial neural networks can also solve problems with complex and nonlinear data, and the neural network models were found to be remarkably close to the experiment results. The values of the experimental results, kinetic coefficients/maximum methane production rate (U_{max}), coefficient of determination R^2/regression R value, mean (SD), root mean square error (RSME)/mean square error (MSE), index of agreement (IA), fractional variance (FV), and predicted results for kinetic studies and AI model are shown in Table 12.

In conclusion, a kinetic model has higher prediction results without taking into consideration various parameters, whereas an AI model is more robust, better at handling information in dynamic conditions, and is able to reduce information overload.

9. Energy Generation from Landfill Leachate Treatment

The global energy demand keeps increasing, which is depleting conventional energy resources, and thus, renewable energy is gaining attention among researchers. Gu et al. [116] reported that the conventional treatment process (AD) of municipal solid waste leachate able to recover of energy up to 37 kWh/m^3.

According to Sonawane et al. [117], Abdoli et al. [118], and Li et al. [119], LFL has a lot of organic and inorganic nutrients that can be utilised in microbial fuel cells (MFCs) for electricity generation. MFC is a bioelectrochemical system and has the capability to generate electrical energy from organic substances in various types of wastewater [120]. This eco-friendly method employs microorganisms as biocatalysts to convert chemical energy in organic waste into direct electric current while treating wastewater [121]. From the results obtained, the greatest open circuit voltage (OCV) of the cell is 1.29 V by using LFL as a substrate for MFCs [117].

Additionally, Abdoli et al. [118] stated that the biogas and methane produced from leachate treatment were 29,897 m^3 and 19,433 m^3 per day, respectively, which can achieve an electrical efficiency of about 40% and requires the capacity of a power plant with 1.8 MW to generate electricity. From the financial analysis, the payback investment period will just take around 1.3 years, with a good internal rate of return equal to 77% or more, as reported by Abdoli et al. [118].

Table 11. Application of conductive materials and conductive nanoparticles in the AD system.

Conductive Materials			Conductive Nanoparticles (CNPs)		
Type	**Concentration**	**Performance**	**Type**	**Concentration**	**Performance**
Bio-based carbon material	• 5 g/L • 1000 cm^2	• Biochar increases methane production rate by 16% [99]. • Carbon cloth increases methane production by 29% [101].	Zero-valent metals	• 1000 mg/L • 5–10 mg/kgVS	• 105.46% biogas production increase by using Fe [100]. • 10% methane formation increase by using Ni [102].
Iron-based	• 25 g/L • 10 g/L	• 34% increase in methane production by using magnetite [103]. • 32% increase in methane production by using magnetite [105].	Metal oxides	• 5, 10, or 20 mg/L • 750 mg/L	• Using Fe_3O_4 will increase the production of biogas by 66% and the formation of methane by 96% [104]. • Methane production goes up by 38% when using Fe_2O_3 [106].

Table 11. *Cont.*

Conductive Materials			Conductive Nanoparticles (CNPs)		
Type	**Concentration**	**Performance**	**Type**	**Concentration**	**Performance**
Carbon-based	• 40 g/L	• Granular activated carbon increases methane generation by 34% [103].	Carbon-based conductive nanoparticles	• 0.5–2 g/L	• 25% of methane yield and 19.5% of biogas production increase by using graphene [107].
	• 200 cm^2 and 12 graphite rods/L	• Graphite increases methane generation by 30–45% [108].		• 1500 mg/L	• 43% methane production increase by using multi-walled carbon nanotubes [109].

Table 12. Kinetic study and Machine learning efficiency in predicting biogas and methane production.

Type of Model	Purpose	Experimental Result	Parameters							Remarks	References
			Kinetic Coefficient/(U_{max})	R^2/Regression R Value	Mean (SD)	RMSE/ MSE	IA	FV	Predicted Results		
First-order model, Stover-Kincannon, Modified Stover-Kincannon, and Van der Meer and Heertjes	First-order model and Stover-Kincannon were used to investigate the kinetics of COD removal via AMBR biological process	Effluent (observed) COD: 1850 mg/L (OLR = 1.04 g COD/L.d), and 25,000 mg/L (OLR = 19.65 g COD/L.d) Mean (SD) for Effluent COD: 11,188 (8644) mg/L	-	R^2 First-order model: 0.926 R^2 Stover-Kincannon: 0.999	First-order model: 9903 (9078) mg/L Stover-Kincannon: 11,025 (8489) mg/L	-	-	-	Predicted COD: First-order model: 1582 mg/L (OLR = 1.04 g COD/L.d), and 27,018 mg/L (OLR = 19.65 g COD/L.d) Stover-Kincannon: 1852 mg/L (OLR = 1.04 g COD/L.d), and 24,038 mg/L (OLR = 19.65 g COD/L.d)	• Stover-Kincannon model showed more consistent output values (R^2) than the first-order model. • Results predicted by the Stover-Kincannon model are much similar to the values measured in the experiments.	[114]

Table 12. *Cont.*

Type of Model	Purpose	Experimental Result	Parameters							Remarks	References
			Kinetic Coefficient/(U_{max})	R^2/Regression R Value	Mean (SD)	RMSE/MSE	IA	FV	Predicted Results		
	Modified Stover-Kincannon and Van der Meer and Heertjes were used to check the kinetic constants of biogas and methane gas production	Biogas: 769 mL/d (OLR = 1.04 g COD/L.d), and 10,470 mL/d (OLR = 18.52 g COD/L.d) Mean (SD) biogas: 4613 (3517) Methane: 423 mL/d (OLR = 1.04 g COD/L.d), and 6177 mL/d (OLR = 18.52 g COD/L.d) Mean (SD) methane: 2705 (2010)	-	R^2 Modified Stover-Kincannon; R^2 Biogas: 0.947907 Methane: 0.934727 R^2 Van der Meer and Heertjes; R^2 Methane: 0.9095	Modified Stover-Kincannon; Biogas: 3845 (3130) Methane: 1928 (1453) Van der Meer and Heertjes; Methane: 2101 (1915)	-	-	-	Predicted biogas and methane: Modified Stover-Kincannon; • Biogas: 848 mL/d (OLR = 1.04 g COD/L.d), and 10,777 mL/d (OLR = 18.52 g COD/L.d) • Methane: 462 mL/d (OLR = 1.04 g COD/L.d), and 5021 (OLR = 18.52 g COD/L.d) Van der Meer and Heertjes; • Methane: 405 mL/d (OLR = 1.04 g COD/L.d), and 6553 mL/d (OLR = 18.52 g COD/L.d)	• Van der Meer and Heertjes model is more suitable for predicting methane production.	-

Table 12. *Cont.*

Type of Model	Purpose	Experimental Result	Parameters							Remarks	References
			Kinetic Coefficient/(U_{max})	R^2/Regression R Value	Mean (SD)	RMSE/ MSE	IA	FV	Predicted Results		
Gompertz model	To predict methane production	Measured Biochemical methane potential (BMP): 78.39 mL/g vs. removed	Umax: 11.28 mL/g vs. removed.d	R^2: 0.994	-	-	-	-	Predicted BMP: 77.98 mL/g vs. removed	• R^2 demonstrated the reliability and accuracy of prediction data. • Thus, the Gompertz model is suitable for predicting the production of methane.	[59]
Fuzzy-based model and Gompertz model	To predict biogas and methane production	-	-	R1 (with nano-ZnO): • R^2 Fuzzy: 0.90 • R^2 Gompertz: 0.77 R2 (without nano-ZnO): • R^2 Fuzzy: 0.90 • R^2 Gompertz: 0.82	-	RSME: R1 (with nano-ZnO): • Fuzzy: 0.15 • Gompertz: 0.16 R2 (without nano-ZnO): • Fuzzy: 0.14 • Gompertz: 0.16	R1 (with nano-ZnO): • Fuzzy: 0.97 • Gompertz: 0.93 R2 (without nano-ZnO): • Fuzzy: 0.97 • Gompertz: 0.95	-	-	• Fuzzy model is more dynamic and robust in terms of predicting methane production since it can consider various parameters, such as input and output variables, compared to the Gompertz model.	[111]

Table 12. *Cont.*

Type of Model	Purpose	Experimental Result	Parameters							Remarks	References
			Kinetic Coefficient/(U_{max})	R^2/Regression R Value	Mean (SD)	RMSE/ MSE	IA	FV	Predicted Results		
Three Layer Back Propagation Artificial Neural Network model (TLBP-ANN)	To determine effective substrate concentration and maximum biogas yield	• Optimum OLR: 16.27 kg COD/m^3 d. • Highest biogas production: 30.07 L/d • COD removal: 89.6%	-	R^2: 0.9703	-	-	0.9882	0.0014	Best linear fit function = 0.9779 experimental + 1.1679, R^2 0.97045	• TLBP-ANN is recognised as the best model for optimising operating parameters, reactor performance, and providing high prediction accuracy. • TLBP-ANN has more accurate and efficient in determining substrate concentration and maximum biogas yield compared to Multiple Nonlinear Regression (MNR) model.	[112]
First-order kinetic (biodegradability) and dynamic activated sludge model (COD removal)	To predict leachate biodegradation and effluent COD in aerobic biological treatment	• Organic load is reduced to 18,950 mg O_2/L COD. • Nitrogen content is reduced to 2319 mg/L. • Ammonia content: 829 mg/L • Nitrate content: 330 mg/L	• Growth coefficient (Y_H) = 0.60 • Half saturation constant (Ks) = 18,950 mg/L • Maximium specific growth rate (μ_H, max)= 0.21/d • *k* aerobic= 0.0146/d • *k* anaerobic= 0.0082/d	-	-	-	-	-	-	• Leachate can be used in agricultural applications, but biological treatment (aerobic) is needed for stabilising fermentation activity and reducing odours. • Effluent COD of leachate can be monitor by a dynamic activated sludge model.	[115]

Table 12. *Cont.*

Type of Model	Purpose	Experimental Result	Parameters							Remarks	References
			Kinetic Coefficient/ (U_{max})	R^2/Regression R Value	Mean (SD)	RMSE/ MSE	IA	FV	Predicted Results		
Artificial neural network (ANN)	To predict biogas production from co-digestion of leachate and pineapple peel	-	-	R Values: • Training: 0.9944 • Validation: 0.9942 • Testing: 0.9800	-	MSE: • Training: 3.95×10^{-2} • Validation: 2.67×10^{-2} • Testing: 1.07×10^{-1}	-	-	• A data separation (% of training set: % of validation set: % of testing set) = 70%: 15%: 15% produced the lowest mean squared error (MSE) for validation. • R values close to 1 were shown, which meant that the neural network model's prediction was in line with the data from the experiments.	• ANN can anticipate biogas production from the ACoD.	[113]

Since fossil resources are limited, Yuvendius et al. [122] revealed that LFL can be used as an alternative electrical generator, which the biogas from organic waste being able to generate 881.6 kW of electricity. Furthermore, LFL can be used as a substrate for bioelectrochemical systems (BES) to generate electricity [123]. BES allows for the recovery of resources from LFL, which include energy, nutrients, metals, and water. However, in the BES treatment of LFL, a suitable pre-treatment and combination with other technologies such as forward osmosis are required in order to achieve maximum resource recovery.

Furthermore, Gu et al. [116] stated that LFL can be used as fertilizer, which has a societal and environmental benefit. Biological treatment, especially AD, is a powerful method for generating energy from organic waste while treating it [124–126]. According to Świechowski et al. [127], the best AD results can be obtained by dividing the process into two stages, which are hydrolysis with acidogenesis, and acetogenesis with methanogenesis. Therefore, it can be concluded that the AD of LFL is a promising resource for the production of renewable energy and the recovery of nutrients and precious materials, with the potential for a high impact on the economy.

10. Limitations and Strategies

There are limited reports in the literature on anaerobic treatment of LFL in pilot and full-scale plants, while most of the studies were conducted in lab-scale. This has led to a lack of comparison of laboratory studies with data from on-site treatment plants. Additionally, the uncertain composition of LFL, complex treatment systems, and the production of ammonia gas and hydrogen sulfide have reduced treatment efficiency. Further research can focus on the purification of methane produced from AD of LFL due to the production of volatile organic compounds (VOCs), including siloxanes, alkanes, terpenes and chlorinated aliphatic hydrocarbons. In commercial-scale applications, the existence of these gases can negatively affect the quality of methane during the conversion process into electricity.

Despite detailed investigations reported by many researchers on AD of LFL, the treated effluent from anaerobic digesters (in single system) has failed to comply with the effluent discharge standard set by EPA and exceeds the allowable limit by about ten-fold. This indicates the urgent need for system optimization, which includes improvement of digester design, modification of HRT and co-digestion with other substrates at optimal mixing ratios.

Many of the research has been conducted in batch studies using artificial LFL with just a handful of studies using genuine FLF in continuous reactor investigations. To comply with the requirements of LFL treatment, future research should be undertaken in various pollutant systems with genuine LFL. Additionally, the review demonstrates certain intrinsic limits of recent developments in AD systems in terms of performance efficiency, payback period, energy production, and the potential to reuse the treated LFL as bio-effluent. Finally, most recent studies have focused on anaerobic treatment systems of LFL without considering techno-economic assessments; thus, information on cost benefit analysis was limited.

11. Conclusions and Recommendations

LFL is a potential substrate for AD and is an economically available source due to its continuous generation via solid waste deposits and composting plants. Publications on AD of LFL in single and integrated systems have increased since 2005. From 2005 to 2022, the integrated system with AD has emerged as an effective system which is more stable with an eco-friendly approach and is more cost-effective compared to other single biological treatments. To improve anaerobic treatment efficiency, pre-treatment methods, co-digestion of LFL with other substrates and integrated systems with AD were investigated extensively. For the pre-treatment of LFL, air stripping achieved the highest COD removal (85%) compared with the coagulation, Fenton and adsorption processes. Additionally, AFBR and AnMBR showed higher COD removal (90%) than other anaerobic reactors. Among the various integrated systems for anaerobic treatment of LFL, the combined process of SBR, PFS coagulation and the Fenton system, and a pair of UBAFs showed a higher COD (97.3%) and ammonia removal efficiency (99%). Additionally, the old LFL achieved higher efficiency as compared to young and intermediate LFL due to the degradation of biopolymers.

Furthermore, during the design and operation of anaerobic system, pH, HRT, organic content (BOD/COD), temperature, OLR, SRT, the effect of moisture content in feedstock, sulphate reduction, the effect of particle size, as well as denitrification should be carefully considered. In addition, there are some potential problems associated with the anaerobic treatment of LFL, which includes a high C/N ratio, extreme salinity content, and a lack of trace element availability in the substrate. The addition of conductive materials (bio-based carbon material, iron-based and carbon-based) and conductive nanoparticles (zero-valent metals, metal oxides, and carbon-based conductive nanoparticles) are proven to be effective in enhancing LFL treatment. The application of kinetic studies and machine learning are excellent predicting tools for biogas production and reactor performance. Digester stability, cost affordability and recycling of energy and materials are other significant characteristics that influence its implementation for the efficient treatment of LFL.

Author Contributions: Conceptualization, N.A.F.Z. and S.I.; validation, S.S. and D.K.; formal analysis, A.M.A.W. and M.A.M.; investigation, A.Z. and D.B.; resources, A.M.A.W. and D.B.; data curation, M.A.M. and A.Z.; writing—original draft preparation, N.A.F.Z. and S.I.; writing—review and editing, D.K., A.Z. and S.S.; visualization, M.A.M., D.K. and D.B.; supervision, S.I. and S.S.; project administration, N.A.F.Z.; funding acquisition, S.I. and A.M.A.W. All authors have read and agreed to the published version of the manuscript.

Funding: This research was financially supported by Universiti Putra Malaysia (UPM), Geran Putra GP-IPS/2022/9713000.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data presented in this study are available on request from the corresponding author.

Acknowledgments: The authors acknowledge the financial support received from UPM GP-IPS/2022/9713000, and Graduate Excellence Programme (GREP), Majlis Amanah Rakyat. Additionally, Master of Water Engineering Program, Department of Civil Engineering and Research Management Centre UPM funded the preparation, writing, and publication of this article. The authors would like to express their gratitude to the top management, Forward Energy Sdn. Bhd toward the success of this study.

Conflicts of Interest: The authors declare no conflict of interest.

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