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REVIEW ARTICLE

Landfill leachate biological treatment: perspective for the aerobic granular sludge technology

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Abstract

Landfill leachates are high-strength complex mixtures containing dissolved organic matter, ammonia, heavy metals, and sulfur species, among others. The problem of leachate treatment has subsisted for some time, but an efficient and cost-effective universal solution capable of ensuring environmental resources protection has not been found. Aerobic granular sludge (AGS) has been considered a promising technology for biological wastewater treatment in recent years. Granules' layered structure, with an aerobic outer layer and an anaerobic/anoxic core, enables the presence of diverse microbial populations without the need for support media, allowing simultaneous removal of different pollutants in a single unit. Besides, its strong and compact arrangement provides higher tolerance to toxic pollutants and the ability to withstand large load fluctuations. Furthermore, its good that settling properties allow high biomass retention and better sludge separation. Nevertheless, AGS-related research has focused on carbon-nitrogen-phosphorus removal, mainly from sanitary sewage. This review aims to summarize and analyze the main findings and problems reported in the literature regarding AGS application to landfill leachate treatment and identify the knowledge gaps for future applications.

Keywords Aerobic granular sludge · Landfill leachate · Biological treatment · High-strength wastewater treatment · Nutrients removal

Introduction

Over the past century, with the fast population growth and rate of urbanization and industrialization, global waste generation has risen significantly to the point where it became one of the world's biggest challenges (Hoornweg et al. 2013). In 2016, the municipal solid waste (MSW) production worldwide was about 2.01 billion tonnes per year, and it is estimated by 2050 that this value will reach 3.40 billion tonnes (Kaza et al. 2018). Despite all the efforts decision-makers devoted to implementing management policies and raising awareness among the population towards more sustainable consumption habits, the increasing waste production will remain a barrier to sustainable development due to the risk it poses to the environment and public health (Environment Agency 2007; European Parliament and Council of the European Union 2008).

Among the different waste disposal methods, landfilling is still the most common worldwide, with around 40% of the total MSW (Kaza et al. 2018), mostly due to its simplicity and relatively lower cost in some regions. Moreover, even if different techniques are employed, there will always be a certain amount of waste that will need to be landfilled, because it either is a by-product without any further use or is the only possible destination for such waste (McDougall et al. 2001).

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One of the consequences of landfilling is the generation of leachate — a high-strength liquid effluent containing a complex mixture of contaminants — that in the absence of proper treatment is a major pollution source, threatening the soil and water sources surrounding the landfill site (Christensen et al. 2001; Environment Agency 2003). Over the years, the leachate characteristics and the different biological and physicochemical treatment processes have been extensively researched and reviewed by several authors (Abbas et al. 2009; Kjeldsen et al. 2002; Lema et al. 1988; Renou et al. 2008; Wiszniowski et al. 2006).

For over a century, biological wastewater treatment by conventional activated sludge (CAS) processes has been used especially due to its good cost-efficiency. However, they usually require high footprint and are very sensitive to abrupt variations of pollutants concentrations (Bengtsson et al. 2019). When treating leachate biologically, some obstacles may impair their efficiency, like the presence of refractory organic matter, high concentrations of ammoniacal nitrogen (NH_3-N), heavy metals, and other toxic inhibitory substances (Renou et al. 2008). For this reason, the development of a robust technology capable of attaining good treatment efficiencies even with highly contaminated wastewaters and complying with the discharge limits imposed by each country is essential.

Among the different biological processes, aerobic granular sludge (AGS) has recently gained much interest in the wastewater treatment field for its unique characteristics that can overcome some of the barriers found in CAS treatment. AGS is a compact and cost-efficient technology characterized by a diverse microbial community capable of carrying out almost all biological conversions in a single system (Gao et al. 2011). Its layered structure allows to, simultaneously, remove carbonaceous organic matter, nutrients (such as nitrogen and phosphorus species), metals, and even some aromatic compounds of difficult biodegradation along the different stages of the operation cycles (Gao et al. 2011; Guo et al. 2020; Sarvajith et al. 2020). Also, granules with compact and denser structures result in faster settling, higher biomass concentration, and lower sludge volumes to be discharded (Guo et al. 2020; Nancharaiah and Reddy 2018).

Considering all its advantages, AGS has been extensively studied and successfully used in domestic wastewater treatment and has also shown promising results for industrial and other high-strength effluents (Abdullah et al. 2011; Corsino et al. 2018; Rosman et al. 2014). For example, Rosman et al. (2014) reported removal efficiencies of 98.4% and 92.7% for chemical oxygen demand (COD) and ammonia, respectively, and 89.5% for total nitrogen in treating rubber wastewater by AGS. In another study reported by de Graaff et al. (2020), mature and stable granules were obtained in the treatment of seawater together with domestic sewage, achieving phosphorus removal values above 90%.

Nevertheless, even though AGS technology emerged just over two decades ago, there are several gaps to be filled regarding its application to high-strength wastewaters such as landfill leachates (Miao et al. 2019; Rani et al. 2020; Ren et al. 2017b, c). This review aims to summarize the state-of-the-art and critically analyze AGS application for the treatment of high-load and recalcitrant wastewaters, focusing on organic matter and nitrogen species removals from landfill leachates. Hence, the main drawbacks and knowledge gaps concerning previous studies of leachate treatment by AGS will be addressed, and some prospects for future applications will also be presented.

Leachate generation and characteristics

Leachate generation

Due to population and economic growth, solid waste generation has exponentially raisen, expected to reach 2.2 billion tons by 2025, according to World Bank forecasts (Iskander et al. 2018). Considering that solid waste disposal in landfills is the most common waste management strategy involving lower costs and low maintenance requirements, attention should be paid to leachate generation since a ton of waste can generate between 0.05 and 0.2 tons of leachate during the stabilization process at the landfill (Wang et al. 2016).

Landfill leachate is a type of wastewater characterized by a high concentration of several pollutants, making this effluent a major threat to the environment and public health. Its generation is mostly related to the infiltration and percolation of water from precipitation and surface runoff through the landfill and to the moisture content of the waste, which, by compression and biochemical reactions, will be released throughout time (Chelliapan et al. 2020; Oller et al. 2011).

Leachate composition

Leachate composition and pollutant load can fluctuate significantly over time. Nonetheless, four main groups of pollutants are often used to characterize leachates (Kjeldsen et al. 2002): (i) organic matter (biodegradable and refractory, like humic and fulvic acids), usually assessed in terms of COD, total organic carbon (TOC), and 5-day biochemical oxygen demand (BOD_5); (ii) inorganic compounds, such as nitrogen compounds (measured as total nitrogen-TN, nitrite-NO_2^-, nitrate-NO_3^-, and ammonia-NH_3), phosphorus (P), chloride (Cl^-), sulfate (SO_4^{2-}) and some sulfide species, carbonate (CO_3^{2-}) and bicarbonate (HCO_3^-), iron (Fe^{2+} and Fe^{3+}), calcium (Ca^{2+}), magnesium (Mg^{2+}), potassium (K^+), and sodium (Na^+), among others; (iii) organic compounds, for instance, polyaromatic hydrocarbons (PAHs), perfluorinated compounds (PFCs), pharmaceuticals, and pesticides; and

(iv) heavy metals. Besides determining these contaminants, other parameters can also be evaluated when characterizing leachates, such as color, turbidity, pH, conductivity, and total and volatile suspended solids (TSS and VSS, respectively), among others.

Inside the landfill, the decomposition of organic matter generates a variety of compounds that will constitute the leachate composition. Humic substances are formed through complex chemical and biological reactions during the humification process, and, due to the covalent bonds of the aliphatic and aromatic fractions, these substances are hydrophilic in the medium (Kjeldsen et al. 2002; Iskander et al. 2018). As a result of the diversity of precursors, types of waste, and environmental conditions, humic substances are macromolecules with very heterogeneous structures, often represented by humic and fulvic acids. In addition, the landfill age greatly influences their formation, with higher concentrations of humic acids being frequently found in older leachates compared to fulvic acids. Some authors also point out that humic substances are resistant to biological degradation, requiring more specific treatments since the multiple redox states increase their recalcitrance (Iskander et al. 2018; Chelliapan et al. 2020).

Impact of different factors in leachate characteristics

Many aspects can contribute to leachate's heterogeneity, such as (i) landfill age, (ii) nature and composition of the waste deposited, (iii) climate conditions and its seasonal variation (mean ambient temperature, precipitation, surface runoff, water permeation, and evaporation), and (iv) landfill characteristics (topography, depth, temperature inside the waste cell, among others) (Bhalla et al. 2013; Gao et al. 2015; Renou et al. 2008).

Since the beginning of its operation, a landfill goes through different stages (aerobic, acetogenic, methanogenic, and stabilization) as the deposited waste decomposes, leading to the formation of distinct types of leachate. Regarding the landfill age, the leachate produced can be categorized as young (<5 years), intermediate (5–10 years), and mature/stabilized (>10 years) (Kjeldsen et al. 2002; Miao et al. 2019; Renou et al. 2008), and its characteristics vary throughout this time (Table 1).

At an early stage of the landfill (aerobic), the oxygen present in the waste interstices is consumed fast and is not renewed as more waste is deposited. The consequent absence of oxygen creates an anaerobic environment that favors the growth of specific microorganisms inside the waste cell, such as hydrolytic, fermentative, and acetogenic bacteria (Kjeldsen et al. 2002).

The hydrolysis of the organic matter and conversion of the resulting amino acids, monosaccharides, fatty acids, and other polymers into carboxylic acids, carbon dioxide (CO_2), and hydrogen (H_2) by fermentative and acetogenic bacteria is the first step of the waste degradation process (Kjeldsen et al. 2002; Lema et al. 1988). This phase generates a leachate with high concentrations of BOD and COD (mostly composed by volatile fat acids — VFAs), higher BOD/COD ratio (increasing leachate's biodegradability), and lower pH values due to the high concentrations of VFAs (Bohdziewicz and Kwarciak 2008; Umar et al. 2010; Wei et al. 2012). Large alkalinity consumption would be necessary to neutralize the acid production and prevent pH drop. If the available alkalinity is not sufficient, the pH decrease may affect the activity of some microorganisms (Ren et al. 2017b). Additionally, although heavy metals concentration in leachates is usually relatively low (Bueno et al. 2020; Robinson 1995), the lower pH values found in these early stages of the landfill increase their solubility into the leachate. Hence, higher concentrations of heavy metals may be reported for young leachates (Christensen et al. 2001; Umar et al. 2010).

Table 1 Leachate composition based on landfill age (Gao et al. 2015; Yadav and Dikshit 2017; Tejera et al. 2019; Meloni et al. 2021; Mojiri et al. 2021)

Parameter	Young	Intermediate	Old
Landfill age (years)	< 5	5–10	> 10
pH (Sørensen's scale)	< 6.5	6.5–7.5	> 7.5
COD (g/L)	> 10	5–10	< 5
BOD_5/COD	0.5–1.0	0.1–0.5	< 0.1
NH_4^+-N (mg/L)	< 400	400	> 400
Heavy metals (mg/L)	> 2	< 2	< 2
Organic species[a]	80% VFA	5–30% VFA + HA + FA	HA + FA
TSS (mg/L)	> 1500	< 1000	< 1000
Landfill stage	Acetogenic	Transition	Methanogenic
Biodegradability	High	Medium	Low

COD chemical oxygen demand, *BOD_5* 5-day biological oxygen demand, *NH_4^+-N* ammonium nitrogen, *TSS* total suspended solids, *VFA* volatile fatty acids, *HA* humic acids, *FA* fulvic acids

[a]Predominant organic compounds for each landfill stage

Over the years, as the landfill matures and enters the methanogenic phase, the conversion of the reaction products from the previous stage into methane (CH_4) and CO_2 by methanogenic microorganisms increases considerably. The consumption of VFAs results in a rise in leachate pH values and a decrease in BOD and COD content, with lower BOD/COD ratios. When the landfill enters the stabilization phase, the CH_4 production rate reaches its maximum and stabilizes for several years, depending on the hydrolysis rate of the organic content present in the landfill (Kjeldsen et al. 2002). Since methanogenic microorganisms predominate in this stage, the fraction of VFAs generated is quickly consumed. Thus, the remaining COD is mainly composed of refractory organic matter, like humic and fulvic acids, which present a great solubility in water (Kjeldsen et al. 2002; Renou et al. 2008; Umar et al. 2010). The COD values usually vary between 500 and 4500 mg O_2/L, and the BOD/COD ratios are normally below 0.1, which is associated with the low biodegradability often found in mature leachates.

Furthermore, the heavy metals' solubility in the leachate is reduced due to the higher pH found in older landfills, which allows the formation of metal precipitates, along with sorption processes on the colloidal matter surface, decreasing leachate toxicity (Iskander et al. 2018; Rani et al. 2020; Wiszniowski et al. 2006). Concerning the nitrogen compounds, ammonia represents a considerable fraction of the total nitrogen present in leachate (Hamza et al. 2019; Miao et al. 2019). Ammonium nitrogen (NH_3-N) is mainly formed via hydrolysis and fermentation of the biodegradable organic matter during proteins degradation. Its concentration tends to increase with landfill age and be very stable under anaerobic conditions (Oliveira et al. 2014; Umar et al. 2010). Contrary to soluble organic substances, the release of nitrogen compounds into the leachate proceeds for an extended period, constituting a problem to biological systems due to its toxicity and inhibitory effect on certain microorganisms (Boonnorat et al. 2018; Ren et al. 2017b).

Microbial communities

In landfills, leachate treatment occurs through several microbial biodegradations and biotransformations of organic and inorganic molecules (Kochling et al. 2015). Therefore, landfill leachate hosts a great diversity of microbial communities, reaching more than 100 different types of genetic sequences and more than 10,000 taxonomic units (Sogin et al. 2006; Meyer-Dombard et al. 2020), presenting a complex taxonomy that helps the biological treatment.

The most abundant taxonomic groups are microbial communities with individuals from the bacterial phyla Firmicutes, Proteobacteria, and Bacteroidetes. Populations of archaea, which typically consist of methanogenic species, are also found (Kochling et al. 2015; Remmas et al. 2017; Song et al. 2015; Zhang et al. 2012). These same authors report that Firmicutes is the dominant phylum in all leachate types, given its dominance and known ability to break down a wide variety of frequently recalcitrant organic compounds. Within the Firmicutes, the dominant class is Clostridia (fermentative acetogens), composed mainly of genera such as Syntrophomonas, Sedimentibacter, Clostridium, and Pelotomaculum (Remmas et al. 2017; Meyer-Dombard et al. 2020). The growth of Sedimentibacter is supported by fermentation of pyruvate or amino acids, while Clostridium uses carbohydrates and/or proteins, depending on the species. The final fermentation products are VFAs (mainly acetate, propionate, and butyrate) and, in the case of Clostridium, also short-chain alcohols and hydrogen. Syntrophomonas is usually found together with methanogenic archaea, with which it syntrophically degrades fatty acids. Song et al. (2015) also point out that *Pseudomonas* (known for degrading recalcitrant organic compounds) is the dominant group in the phylum Proteobacteria, reaching 92.4% of the total abundance.

Importantly, the decomposition stage and landfill age greatly affect the microbial community structure. As the landfill becomes older, biodiversity increases, presenting a diverse specialized bacterial community capable of degrading recalcitrant organic compounds and resisting the high concentrations of heavy metals accumulated in this effluent (Remmas et al. 2017). With the increasing age of the landfill, the abundance and diversity of the phylum Firmicutes also increase. The abundance of the phylum Proteobacteria decreases, giving rise to the phylum Spirochaetes, which also becomes dominant in this type of leachate. The Bacteroidetes count does not show a linear trend over time (Kochling et al. 2015).

In particular, old landfill leachate can serve as a reservoir for isolating specialized degrading bacteria, which can be used in the bioremediation and bioaugmentation of toxic compounds accumulated in contaminated soils and aquatic environments. For example, strains of *Pusillimonas* were involved in the bioremediation of aged soils polluted with creosote (Lladó et al. 2013; Remmas et al. 2017).

Landfill leachate treatment

Selection of the treatment process

In virtue of leachate's high pollutant load, applying a treatment strategy before disposal is mandatory, which can either be performed externally (off-site treatment) or on the landfill site (on-site treatment) (Figure 1).

For many years, the off-site co-treatment of leachate with municipal sewage in wastewater treatment plants (WWTPs) was very common due to its simplicity, smaller investment in structures, and lower operation/maintenance costs (Campos et al. 2019; Renou et al. 2008). In addition, this mixture reduces leachate toxicity through its dilution with domestic wastewater and may also increase biodegradability by balancing the carbon-nitrogen-phosphorus ratio (Dereli et al. 2021; Ferraz et al. 2016; Gao et al. 2015). However, some disadvantages have contested this strategy's effectiveness, like the high loads of slowly biodegradable organic compounds, ammonium nitrogen, heavy metals, and other inhibitory substances to the biological processes of the municipal WWTPs, hindering the compliance with the discharge limits (Dereli et al. 2021).

Nonetheless, for on-site treatment, the operation and maintenance of a leachate treatment plant (LTP) on the

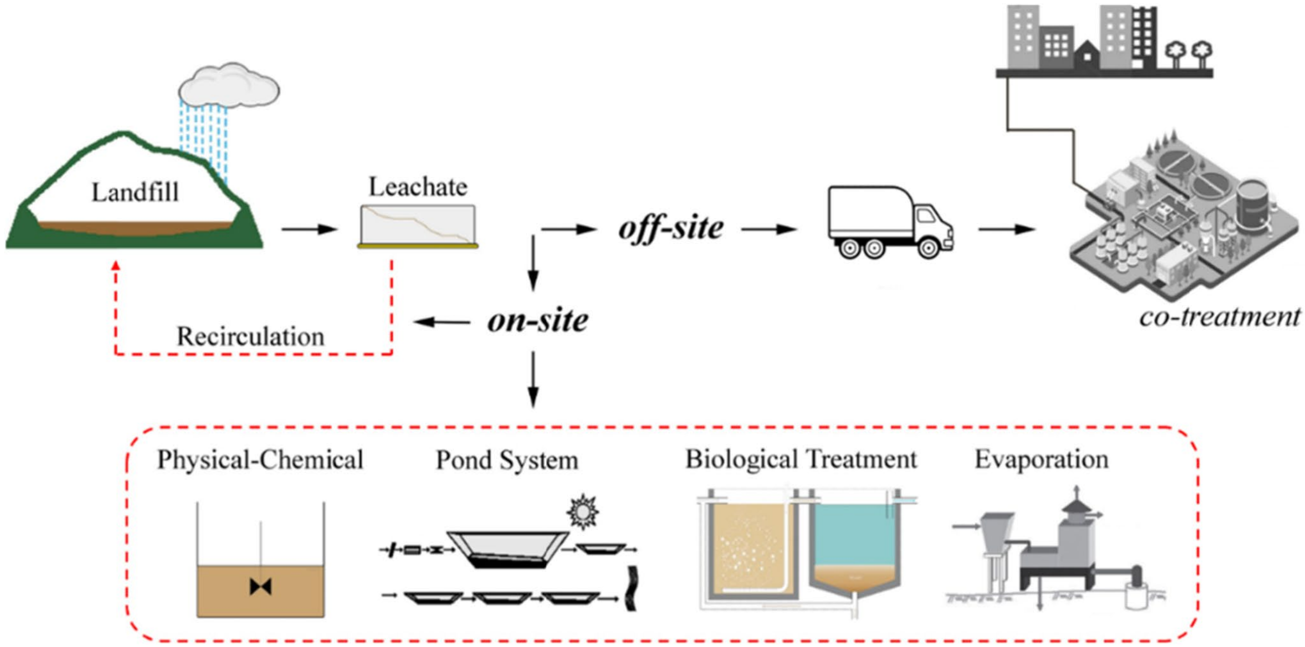

Figure 1 Off-site and on-site leachate treatment processes

landfill location is required, which can combine different physical, chemical, and biological processes (Campos et al. 2019; Ferraz et al. 2016; Yuan et al. 2016). The variability in leachate composition with landfill aging, like the increase in ammonia content and lower COD (mainly refractory organics), not only increases leachate toxicity but also creates nutrient imbalances that can significantly impair its biological treatment, making the conventional treatment methods less efficient (Brennan et al. 2017; Wu et al. 2015). It has been reported that in high concentrations, free ammonia (FA) and free nitrous acid (FNA) can strongly inhibit the activity of ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB), reducing the effectiveness of the nitrification process (Ferraz et al. 2016; Soliman and Eldyasti 2018; Wang et al. 2016; Yang et al. 2004). Additionally, the lack of biodegradable organic carbon sources can also harm denitrification, which leads to lower nitrogen removal efficiencies (Miao et al. 2014; Peng et al. 2008). In such cases, the external addition of nutrients and readily biodegradable carbon sources to the biological process is often used to improve the carbon and nitrogen removal from leachate, consequently raising the treatment costs (Dereli et al. 2021; Zhao et al. 2012). Therefore, adding complementary treatment steps to remove recalcitrant compounds becomes necessary (Di Iaconi et al. 2006).

Physicochemical processes

Leachate treatment by physicochemical processes is commonly used as a complementary step to biological treatment. Particularly in older leachates, where BOD_5/COD ratio is very low and refractory organics are predominant, the biological treatment is usually insufficient to achieve the desired characteristics for discharge. Thus, some physical and chemical processes can be included in the treatment strategy, either as a pre-treatment to eliminate target compounds that inhibit the following biological processes or as a final polishing step to remove the remaining recalcitrant organic matter and some toxic substances (Renou et al. 2008).

Several researchers have studied in the past different physical and chemical processes, such as flotation (Palaniandy et al. 2010; Zouboulis et al. 2003), coagulation/flocculation (CF) (Amokrane et al. 1997; Tatsi et al. 2003; Zamora et al. 2000), chemical precipitation (Altinbaş et al. 2002; Calli et al. 2005; Li et al. 1999), chemical oxidation/advanced oxidation processes (AOPs) (Chen et al. 2019; Oulego et al. 2016; Silva et al. 2016; Zhang et al. 2005), adsorption onto activated carbon (Imai et al. 1995; Kargi and Pamukoglu 2003; Morawe et al. 1995; Rodríguez et al. 2004), ion exchange (Boyer et al. 2011; Fernández et al. 2005), and membrane filtration (Marttinen et al. 2002; Pirbazari et al. 1996). The combination of different methods (physicochemical and biological) has proven to be the most efficient way to remove both recalcitrant organic matter (shown by the low COD values after treatment) and ammonium nitrogen from stabilized leachates (Kurniawan et al. 2006; Marttinen et al. 2002). It is noteworthy that this choice must always consider the initial leachate characteristics.

However, the use of physicochemical processes can also have some disadvantages, like the higher costs associated with energy consumption and chemicals addition to the system and the production of high volumes of sludge and subsequent need for its treatment/disposal. The limited applicability and chance of toxic by-product formation have also been reported as an inconvenience (Kurniawan et al. 2006).

Biological processes

Conventional activated sludge systems are, among biological processes, the most commonly used in leachate treatment. As previously mentioned, these systems often come across a few obstacles related to recalcitrant organic matter and nutrient removal (Chys et al. 2015). The presence of toxic aromatic compounds, large ammonium nitrogen concentrations, and high salinity environments not only can significantly inhibit AOBs and NOBs and compromise the nitrification process but may also negatively affect the biomass settling properties (Deng et al. 2018; Park and Bae 2009; Ramos et al. 2015).

Additionally, the secondary clarifiers in CAS systems are often affected by sludge bulking due to the excessive growth of filamentous bacteria, resulting in poor settleability, low sludge compaction, and high concentrations of suspended solids in the treated effluent (Li et al. 2011; Ren et al. 2017b; Zou et al. 2019). Sludge bulking can occur when the food to microorganisms (F/M) ratio and dissolved oxygen concentrations are low but may also be a consequence of high concentrations of sulfide, oils and greases, or when organic substrates are rapidly metabolized (Li et al. 2011; Martins et al. 2004; Zou et al. 2019). Consequently, there will be a reduction in the abundance of slow-growing microorganisms, usually nitrifying or biodegrading microorganisms with low growth kinetics. Likewise, during the denitrification process, the conversion of the high nitrogen content (resulted from nitrite and/or nitrate accumulation) into gaseous nitrogen (N_2) can lead to sludge loss and formation of scum on the secondary clarifier surface and in the anoxic zones of the aeration tank (Zhang et al. 2017).

Besides CAS systems, other technologies have been reported in the literature regarding biological leachate treatment, such as sequencing batch reactor (SBR) (Uygur and Kargı 2004), membrane bioreactor (MBR) (Xue et al. 2015), moving bed biofilm reactor (MBBR) (Chen et al. 2008), fluidized-bed biofilm reactor (FBBR) (Eldyasti et al. 2010), rotating biological contactor (RBC) (Castillo et al. 2007), trickling filter (TF) (Matthews et al. 2009), and also anaerobic systems like anaerobic filter (AF) (Wang and Banks 2007) and up-flow anaerobic sludge blanket (UASB) reactor (Castillo et al. 2007), among others. However, most of the difficulties noticed in CAS processes were also found in these systems (Deng et al. 2018; Peng et al. 2018).

As an alternative, AGS technology can tolerate high pollutant loads in the influent and still achieve COD, TN, and total phosphorus (P_{Total}) removals above 90% (Nancharaiah et al. 2018). Most of the microbial groups in aerobic granules are resistant to the toxic compounds present in the leachate without compromising their performance and granules' stability (Ren et al. 2017a, c). In addition, when compared to CAS systems, AGS has lower associated costs (less 20–25% in operation and 23–40% in energy consumption) and lower footprint (50–75% lower) (Bay Area Clean Water Agencies 2017; Bengtsson et al. 2019). Therefore, AGS systems emerge as a promising technology to replace obsolete conventional biological systems.

Despite being an excellent alternative for the treatment of domestic and industrial wastewaters, further investigations are required with landfill leachates, such as the need for dilution, physicochemical pre-treatment, external carbon source addition, and different cycle times, among others. Additionally, pilot-scale studies for medium and long-term evaluation of leachate effect on the aerobic granule properties and efficiency of simultaneous removal of pollutants are needed.

Aerobic granular sludge technology in leachate treatment

Aerobic granules consist of dense spherically shaped aggregates of microorganisms bounded through physical, chemical, and biological phenomena (Liu and Tay 2004). Their large size and compact structure provide exceptional settling abilities and great water-sludge separation, producing lower and very concentrated sludge volumes, thus eliminating the need for a secondary clarifier (Franca et al. 2018).

The main advantages of AGS compared to other biological systems are the following: (i) retention of high biomass concentrations in the bioreactor, (ii) presence of different redox microenvironments (anaerobic, anoxic, and aerobic regions) due to its layered structure, (iii) possibility of controlling different metabolic reactions through the adjustment of dissolved oxygen concentrations, (iv) metabolic cooperation between autotrophic and heterotrophic microorganisms, and (v) capacity to withstand high influent loads and hydraulic shocks (Gao et al. 2011; Nancharaiah and Reddy 2018; Rosman et al. 2014; Zheng et al. 2020).

Several studies have shown that AGS is capable of treating high-strength effluents containing large concentrations of ammonia (Wei et al. 2012), organic matter (Xiong et al. 2020), phosphorus (de Graaff et al. 2020), and even aromatic compounds (Ramos et al. 2015), being an interesting biological alternative for leachate treatment (Ren et al. 2018).

Even though AGS technology has been applied in the most diverse wastewater treatment fields, the number of studies using this technology in leachate treatment is relatively small. So far, advances in the application of AGS to treat leachate can be divided into three main phases: (i) investigation of the need for pre-treatment and optimization of the AGS reactor aeration system (2012–2014), (ii) study of different dilutions of leachate influent to the AGS and comparative analysis with activated sludge reactors (2014 to 2017), and (iii) leachate co-treatment with domestic sewage (2017 to 2020).

Wei et al. (2012) carried out one of the first studies using aerobic granules to treat municipal landfill leachate, with and without pre-treatment for NH_3-N removal. The pre-treatment favored the granulation process since the high NH_3-N concentrations impaired nitrogen and COD removals. On the other hand, in work reported by Di Bella and Torregrossa (2014), the nitrogen removal was satisfactory without any pre-treatment due to the acclimatization period of the granules with leachate during its cultivation. This is very important for selecting specific/specialized microorganisms to degrade the compounds present.

The progress on studies applying aerobic granular biomass in leachate treatment has allowed the comparison of performance with other systems, especially with CAS. The superiority of AGS over CAS was evidenced, both in COD and nitrogen removals, in addition to granules being less sensitive to high loads and toxicity (Ren et al. 2017a, b, 2018).

The third phase of studies evaluated the possibility of co-treating leachate with domestic wastewater, admitting leachate proportions between 20 and 60% (Ren et al. 2017c). This mixture significantly benefited the carbon and nitrogen removals, but the efficiency decayed considerably for higher pollutant concentrations (Bueno et al. 2020). The lack of acclimatization period or higher leachate ratios may decrease the mixed liquor suspended solids (MLSS) concentrations due to granules' disintegration and biomass washout (Table 2). When the solids' loss is superior to biomass

Table 2 Biomass retention in the reactor: effect of the granulation process and type of leachate treated in the concentration of mixed liquor suspended solids

Leachate type	Granulation process	Initial MLSS[a] (mg/L)	Final MLSS[a] (mg/L)	References
Leachate pre-treated for NH_3-N removal	Leachate + PAC [b)]	4000	3116	Wei et al. (2012)
Leachate without pre-treatment	Leachate + PAC [b)]	4000	3083	
Real leachate diluted with tap water up to a COD of 9700 mg/L	Synthetic wastewater (with lower pollutant loads)	11,000	< 5000	Di Bella and Torregrossa (2014)
Leachate diluted with synthetic wastewater up to a COD of 4500 mg/L	Synthetic wastewater (with lower pollutant loads)	11,000	< 5000	
Synthetic old leachate[c]	Synthetic old leachate[c]	8070	8070	Ren et al. (2017a)
Synthetic young leachate	Synthetic young leachate	< 4000	6943	Ren et al. (2017b)
Leachate diluted with municipal wastewater (leachate volume ratio 10–40%)	Municipal wastewater	3215	6000	Ren et al. (2017c)
Leachate diluted with municipal wastewater (leachate volume ratio: 60%)	Municipal wastewater	6000	10,000	
Leachate diluted with municipal wastewater (leachate volume ratio: 90%)	Municipal wastewater	10,000	9000	
Leachate diluted with municipal wastewater (leachate volume ratio:10–65%)	Leachate (ratio: 10%) diluted with municipal wastewater	2591	6476	Ren et al. (2018)
Leachate diluted with municipal wastewater (leachate volume ratio: 65–90%)	Leachate (ratio: 10%) diluted with municipal wastewater	6476	14,533	
Leachate diluted with municipal wastewater (leachate volume ratio: 90–100%)	Leachate (ratio: 10%) diluted with municipal wastewater	14,533	12,707	
Leachate diluted with synthetic wastewater: 5%	Leachate + synthetic wastewater (low pollutant load)[d]	3325	1525	Bueno et al. (2020)
Leachate diluted with synthetic wastewater: 10%	Leachate + synthetic wastewater (low pollutant load)[d]	1525	2695	
Leachate diluted with synthetic wastewater: 20%	Leachate + synthetic wastewater (low pollutant load)[d]	2695	2776	

[a]MLSS — mixed liquor suspended solids; Initial MLSS: value before granulation; Final MLSS: value after the leachate treatment by AGS

[b]PAC — powder activated carbon

[c]Tannic acid used to simulate the refractory organic matter

[d]Acclimatization period (40 days) only with synthetic wastewater and then granulation simultaneously with the treatment (leachate diluted in the synthetic wastewater, increasing the leachate volume ratios)

growth, a decrease in MLSS concentration is observed, as previously reported (Bueno et al. 2020; Di Bella and Torregrossa 2014). Therefore, the composition of the leachate influent to the system has a significant impact on the granulation process.

Some parameters are essential to evaluate AGS reactor operation and control treatment efficiency, such as sludge retention time, dissolved oxygen concentrations, cycle duration, and settling phase (Table 3). They will influence the granules' formation, structure, stability, the bacteria distribution inside the granule, and their metabolic reactions (Franca et al. 2018). Nevertheless, the use of SBR reactors to treat leachate with AGS technology still presents many gaps to be filled, requiring further studies.

Formation and maintenance of the granules

One of the main differences between AGS and other types of biomass is the higher extracellular polymeric substances (EPS) production (Nancharaiah and Reddy 2018; Sun et al. 2015). The formation of stable and mature granules will depend highly on the operating conditions and environmental stress they are subjected to (Franca et al. 2018).

Aerobic granules are usually produced using activated sludge for the reactor startup. Upon the first contact with the leachate, the sludge in the AGS system starts to present a flocculating structure and dark brown color (Bueno et al. 2020; Wei et al. 2012). The time necessary for granulation to occur will depend on the influent characteristics. In other words, high carbon and nitrogen loads can significantly delay granule formation (Szabó et al. 2016).

Bueno et al. (2020) reported granules formation in the first 40 days of reactor operation after the acclimatization phase. During this period, the initial flocs in biomass went from a diameter of 10–95 µm to the formation of granules with a diameter of 382–421 µm in 45% of the biomass. At high loads, irregularities in the surface and structure of the granules are common, even after process stability. Thus, after the leachate incorporation, it has been observed that within 60 days, the biomass presented good aggregation, excellent settling properties, and a majority of irregular granules (Ren et al. 2017b, 2018; Wei et al. 2012). The proportion of leachate diluted with domestic wastewater also influences the size of the granules, i.e., higher leachate ratios produce smaller granules and vice versa (Bueno et al. 2020; Ren et al. 2017c).

Table 3 Main operating parameters of AGS reactors reported on literature for leachate treatment (Bueno et al. 2020; Di Bella and Torregrossa 2014; Ren et al. 2017c; Ren et al. 2018; Wei et al. 2012)

Parameters	Reference value
Leachate dilution	10–60%
Concentration of ammoniacal nitrogen	< 788 mg NH_3-N/L
Concentration of solids in the reactor	> 3 g TSS/L
Cycle time	12–24 h
SRT	< 30 d
Dissolved oxygen	2–4 mgO_2/L
Settling time	< 10 min
Volumetric exchange ratio	50%
SVI_5/SVI_{30}	1.2–1.8

SRT sludge retention time, SVI_5 sludge volume index (after 5 min of settling), SVI_{30} sludge volume index (after 30 min of settling)

Special attention should be paid between the 3rd and 4th months of operation. During this period, the selection pressure can still eliminate the sludge of worse quality that presents problems of sedimentability and difficulty to create anoxic/anaerobic zones inside the granule, causing TSS loss in the treated effluent. In addition, biomass disintegrates more easily, especially at concentrations approximately above 200 mg NH_3-N/L (Ren et al. 2017c). However, even after observing that lower leachate concentrations favor granules' stability, Bueno et al. (2020) reported a significant TSS loss for the lowest leachate ratios (5 and 10%). Therefore, TSS loss during the granulation process may not depend on the leachate proportion used but instead be a consequence of the natural sludge selection occurring inside the reactors, where the less quality sludge is discarded (Ren et al. 2017a, b).

Only after 90 days of reactor operation that 80–90% of the biomass was granular, with average sizes of 480–612 µm, showing good stability and without uncontrolled sludge losses (Bueno et al. 2020; Ren et al. 2018). After this period, the granules showed insignificant growth. However, controlling the organic load after the stabilization period is very important, as significant changes can affect granules' integrity (Di Bella and Torregrossa 2014).

Sludge features in AGS systems for leachate treatment

When treating wastewater with high pollutant loads, it has been observed that parameters such as sludge volume index (SVI) and TSS present a slightly different behavior than expected for conventional loads, meaning that by increasing the COD and ammonia concentrations fed to the system, SVI tends to decrease (Abdullah et al. 2011; de Kreuk and van Loosdrecht 2004; Kocaturk and Erguder 2015; Sarvajith et al. 2020; Xiong et al. 2020). Some studies point out that the SVI_8/SVI_{30} or SVI_5/SVI_{30} ratio can be considered a good predictor of granulation in waters with high pollutant loads. In other words, a ratio: (i) above 1.8 indicates the thickening of the sludge blanket, (ii) between 1.2 and 1.8 indicates the predominance of aerobic granules in the biomass, and (iii) closer to 1.0 shows that the sludge majority is constituted by granules (Corsino et al. 2018; de Kreuk et al. 2005; Hamza et al. 2018; Kocaturk and Erguder 2015; Ni and Yu 2010; Schwarzenbeck et al. 2004; Yilmaz et al. 2008).

Of the few studies reported in the literature regarding leachate treatment by AGS, only some monitored SVI_{30} and SVI_5 after granulation and during the process. Therefore, it is not possible to establish a clear pattern regarding the contaminant loads and their effect on the SVI. However, there is a consensus that the sedimentation velocity increases when the proportion of leachate incorporated into the system increases (Bueno et al. 2020; Ren et al. 2017b).

According to Ren et al. (2017b), SVIs lower than 50 are acceptable in treating effluents with high loads and guarantee a good sludge sedimentation. In most works, granulation was obtained through low-load synthetic effluents or with low leachate proportions (Table 2). It appears that when the leachate is incorporated into the process or its proportion increases, the SVI decays (Table 4), possibly due to the granulation optimization or the frequent washouts and granules' disintegration.

In the reported studies, most MLSS are volatile and with very different concentrations (Table 4). Depending on the leachate proportion fed to the system, they may decrease (Bueno et al. 2020; Di Bella and Torregrossa 2014; Wei et al. 2012) or increase with the increase in leachate ratio (Ren et al. 2017b, c, 2018). In addition, very high MLSS concentrations (about 8 g/L) were achieved in some studies (Ren et al. 2017c, 2018), while others did not surpass 4 g/L (Bueno et al. 2020; Ren et al. 2017b).

Solids loss from the mixed liquor is due to the increase in the NH_3-N load, which can negatively affect the denitrification and lead to biomass washout from the system (Bueno et al. 2020; Di Bella and Torregrossa 2014; Ren et al. 2017b). Also, high leachate loads reduce cellular hydrophobicity and directly affect sludge aggregation, forming a flocculent sludge with a higher possibility of being discarded with the effluent (Ren et al. 2017b, c).

Therefore, the inoculum quality is important in the recovery after loads shock. A good quality inoculum allows the system to stabilize quickly after the sludge loss period and favors high biomass growth and VSS concentrations (Ren et al. 2017c). However, to minimize the excessive loss of solids, Bueno et al. (2020) suggested that some strategies can be used in these systems, such as incorporating a secondary sedimentation tank after the AGS reactor.

In addition, high sedimentation velocities in mixed liquor are crucial for increasing solids concentration at significant levels. Ren et al. (2018) related the MLSS concentration to the sludge age and found that the older the sludge, the greater the MLSS concentration. However, if the sludge age is too high (for example, above 30 to 40 days), the MLSS concentration will decrease, although the concentration of VSS may still increase.

Optimization of leachate treatment by AGS

Pre-treatment

As previously mentioned, despite presenting numerous advantages for leachate treatment, mainly regarding implementation and operational costs, the removal of nutrients (especially nitrogen) and recalcitrant compounds can be very low in biological systems. In turn, physicochemical processes have been widely used to reduce part of the influent nitrogen load from biological treatment systems or to remove recalcitrant compounds, although few studies refer to the removal of toxicity from the final effluent (Di Bella and Torregrossa 2014; Oulego et al. 2015; Queiroz et al. 2011).

Previous works have used gradual dilutions, coagulants/flocculants, physical processes of separation by gravity, conventional fat removal processes, and even a previous biological treatment as leachate pre-treatment (Corsino et al. 2017; Kocaturk and Erguder 2015; Świątczak and

Table 4 Physical parameters reported in the literature before and after adding leachate

Leachate dilution (%)	After granulation				End of the process				References
	TSS (mg/L)	VSS (%)	SVI_5 (mL/g)	SVI_{30} (mL/g)	TSS (mg/L)	VSS (mg/L)	SVI_5 (mL/g)	SVI_{30} (mL/g)	
5	5420	86	400	210	3325	2835	276	155	Bueno et al. (2020)
10	3325	85	62	155	2695	2319	77	62	
20	2695	86	36	77	2776	2085	36	30	
10–40	3215	85	45	40	6000	5300	20–25	15–20	Ren et al. (2017c)
60	6000	88	20–25	15–20	9500	8800	20	15	
90	9500	92	20	15	9900	7900	25	20–25	
10–65	6476	87	19	---	7500	5000	19	---	Ren et al. (2018)
65–90	7500	66	19	---	14,533	8633	19	---	
100	14,533	59	19	---	12,707	7878	19	---	

TSS total suspended solids, *VSS* volatile suspended solids, *SVI_5* sludge volume index (after 5 min of settling), *SVI_{30}* sludge volume index (after 30 min of settling)

Cydzik-Kwiatkowska 2018). Wei et al. (2012) applied magnesium oxide and phosphoric acid coupling to struvite precipitation, resulting in a larger proportion of granules' growth and higher efficiencies of simultaneous nitrification and denitrification (SND). Their main goal was to remove NH_3-N during the pre-treatment, especially FA that is considered toxic to the process. Although the retained sludge is conducive to soil fertilization, controlling its production and disposal is necessary. Attention should also be paid to the precipitant required dosage and the process sensitivity to pH (Kurniawan et al. 2006).

Coagulation/flocculation satisfactorily reduces the levels of adsorbable halogenated organic compounds, suspended solids, heavy metals, polychlorinated biphenyls, and humic substances such as humic and fulvic acids. Organic colloidal compounds are thermodynamically stable, presenting negative surface charges. Thus, coagulation consists of particles destabilization by neutralizing surface electrical forces and reducing the repulsive forces between them, while flocculation aimed to increase the volume and density of the particles, which can be removed by sedimentation or flotation (Miao et al. 2019; Rani et al. 2020; Yuan et al. 2016).

As the process efficiency depends on the molecular weight of the organic particles to be removed, CF has been more suitable for old landfills. The most frequently used coagulants are ferric chloride and aluminum sulfate due to their excellent cost-efficiency ratios. However, coagulants can reduce the final effluent quality by increasing the concentration of iron, aluminum, chloride, or sulfate (Rui et al. 2012). Since there is no NH_3-N removal and leachate biodegradability is virtually unaffected, CF is commonly combined with other processes (Rui et al. 2012; Torretta et al. 2017).

In addition to CF and chemical precipitation, adsorption has been widely used when the goal is to remove recalcitrant and non-biodegradable organic compounds, with COD removal efficiencies over 90%. The most commonly used adsorbent is activated carbon, which efficiently removes carbon, metals, and other compounds but does not remove ammoniacal nitrogen. Still, activated carbon regeneration requires large energy consumption, which implies higher costs (Campos et al. 2019; Li et al. 2019; Miao et al. 2019).

Alternatively to biological nitrification, ammonia stripping has been used for NH_3-N removal, eliminating volatile organic compounds as well. Mass transfer to the gas phase occurs for some constituents due to the large volume of air injected into the leachate, in which the increase in temperature and pH also favors the process (Gao et al. 2015). Therefore, the exhaust gas must be properly controlled and undergo adequate treatment before being discharged into the atmosphere, thus avoiding air pollution and the release of toxic volatile compounds.

In general, besides favoring nitrification and biological denitrification, physicochemical pre-treatment makes the process more efficient and less toxic. However, due to the complex composition of the leachate, there is a need for combining different treatment strategies since there is not a single system that can remove all pollutants present in the leachate. Gomes et al. (2019) proposed a treatment in multiple stages combining biological processes with a physicochemical treatment and an advanced oxidation technology (AOT) (Figure 2). The first stage of the overall process took place in SBRs with 24-h cycles so that TN and alkalinity reached values below 15 mg N/L and 1.1 g $CaCO_3$/L, respectively. In line with the multistage system reported by Silva et al. (2017), coagulation had the same effect by precipitating humic acids and removing colloidal and suspended material, increasing the photo-based post-treatment efficiency. Lastly, the final biological oxidation guaranteed compliance with the legal COD and TSS levels imposed by the legislation in force.

Accordingly, due to the leachate refractory character and the high loads of organic matter and nitrogen, the integration of physicochemical processes with biological oxidation has proved to be an excellent alternative. The association of both types of technologies, in addition to reducing the concentration of organic and nitrogen species, also removes humic and fulvic acids, which present low removals in isolated biological processes. Moreover, the leachate pre-treatment in AGS systems should occur using coagulants that do not cause

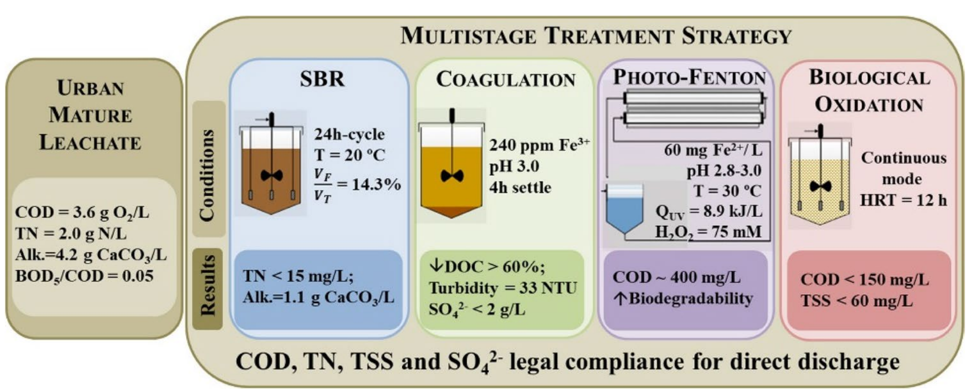

Figure 2 Multistage treatment combining biological and physicochemical processes in the treatment of an urban mature leachate (Gomes et al. 2019)

sudden changes in pH, such as aluminum-based coagulants (Rui et al. 2012; Wiszniowski et al. 2006). Nevertheless, there is still a lot to explore regarding combining AGS reactors with physicochemical systems, especially in pilot- or full-scale applications.

Post-treatment

When treating a young to intermediate leachate, the efficiency of the physicochemical treatments integrated with biological treatment has been satisfactory, minimizing the disadvantages of each process alone. However, with the aging of landfills, these conventional treatments (physical-chemical-biological) are not sufficient to achieve environmental compliance, so other approaches must be applied. AOTs have been proposed in recent years as an effective alternative for the oxidation of bio-refractory organic compounds from landfill leachate into biodegradable organic compounds or even its total mineralization into CO_2, H_2O, and inorganic compounds (Luo et al. 2020).

These processes include the following: (i) non-photochemical methods, such as ozonation (O_3), perozonation (O_3/H_2O_2), catalytic ozonation (O_3/catalyst), and Fenton processes (H_2O_2/Fe^{2+}), or (ii) photochemical methods, for instance, O_3/UV, H_2O_2/UV, O_3/H_2O_2/UV, photo-Fenton (Fe^{2+}/H_2O_2/UV-Vis), and photocatalysis (UV/catalyst) (Costa et al. 2019; Wiszniowski et al. 2006). In addition, electrochemical methods can be used, such as the Fenton process combined with an electrochemically generated oxidizing agent and catalyst. However, AOTs are usually expensive processes, requiring high doses of oxidants and efficient control systems, and are energy demanding. In this regard, among the existing AOTs, Fenton- and ozone-based processes are the most used methods for leachate treatment (Bassam et al. 2012; Leszczyński and Maria 2018).

Septiariva et al. (2019) employed ozonation as a post-treatment of old leachate, increasing COD removals from 51 to 65% compared to the isolated biological process, while in young leachate, there was no significant difference. Mokhtarani et al. (2014) also evaluated the performance of an ozone post-treatment on a biologically pre-treated leachate featuring COD values between 0.5 and 1 g O_2/L and obtained COD removals of 56% (at pH 9, ozone dose of 0.4 g O_3/h and 60 min of reaction). Furthermore, Soubh and Mokhtarani (2016) studied the combination of O_3 and sodium persulfate as a post-treatment method, reaching 84% COD removal after 210 min of reaction in the optimal conditions (at pH 9, ozone dose of 0.79 g O_3/h and 4.5 g/L sodium persulfate). Besides, the combined process (O_3/persulfate) resulted in lower ozone consumption rates compared to ozonation alone (0.35 and 1.16 mg O_3/mg COD removed, respectively).

Mahdad et al. (2016) compared the post-treatment by the conventional Fenton and the photo-Fenton processes. The Fenton process removed 55.9% of COD and 65.7% of color, while the photo-Fenton treatment reduced 73.8% of COD and 83.6% of color and increased the effluent biodegradability. Notwithstanding, by allowing the process to occur close to neutrality, ozonation becomes a more promising post-treatment strategy for biological reactors, while Fenton processes require acidic pH values (Roy et al. 2018; Wiszniowski et al. 2006).

Cycle time

In AGS reactors, nitrogen and phosphorous removal mechanisms occur throughout the entire operating cycle. After the feeding stage, the reaction begins, and it can be the following: (i) entirely aerobic, (ii) aerobic and anaerobic, (iii) aerobic and anoxic, or (iv) other combinations. Most SBRs used for AGS cultivation in conventional loads operate with 4 to 12 h cycle time. However, depending on the influent load, longer cycle times can favor the granulation process and improve AGS efficiency. Di Bella and Torregrossa (2014) indicated that for high-strength effluents such as leachate from landfills, the cycle time should not be lower than 12 h. Other authors also reported that short cycles are not enough to achieve high removal efficiencies and favor the aerobic biomass characteristics in terms of EPS content and settling capacity, among others (Bueno et al. 2020; Ren et al. 2017a, 2018; Wei et al. 2012). Additionally, short cycles (for example, lower than 4 h) can cause VSS loss through biomass washout but cycles greater than 24 h are subject to the absence of nitrifying granulation (Nancharaiah and Reddy 2018).

The alternation of aerobic and anaerobic conditions favors the growth of microorganisms beneficial to granulation (phosphorus-accumulating organisms-PAOs, glycogen-accumulating organisms-GAOs, nitrifying bacteria) since the proliferation of heterotrophic microorganisms is suppressed by the lack of a soluble carbon source under aerobic conditions (Rollemberg et al. 2018). However, since phosphorus concentration is not significant in the leachate, an extended anaerobic feeding may be enough to select PAOs and GAOs (Bueno et al. 2020; Ren et al. 2017b).

The selection of microorganisms in the system is also dependent on the sedimentation time, which is a key factor in the formation of aerobic granules. The SBR operation with a short settling time (< 10 min) allows for a quick assortment of microbial aggregates by creating a high selective pressure, causing the washing of lighter microbial flocs and favoring granulation by creating a relatively high shear force (Nancharaiah and Reddy 2018; Yuan et al. 2019). All the studies applying AGS to treat leachate had a settling time of 5 min in an attempt to obtain a more stable and consistent granulation (Table 4).

Anaerobic and staggered feeding

According to de Kreuk et al. (2005), in addition to organic load, shear force, selection pressure, and substrate composition, among others, the feeding mode also affects the formation and stability of aerobic granules.

After pre-treatment, the anaerobic feeding is considered essential for the granules formed to remain stable and with good activity and for phosphorus removal by phosphate-accumulating microorganisms (PAOs) (de Kreuk and van Loosdrecht 2004; Hamza et al. 2018). During the anaerobic feeding period, all acetate is converted into internal storage polymers (e.g., polyhydroxybutyrate — PHB), and the phosphate is released into the liquid. Then, in the aerobic period, there is cell growth from the stored PHB and intracellular conversion of the phosphate available in the liquid phase into the polyphosphates. Thus, the selection of these organisms resulted in smooth, dense, and stable granules (de Kreuk and van Loosdrecht 2004).

This strategy seems to be very well accepted and widespread when using AGS to treat leachate. Except for Di Bella and Torregrossa (2014), all other studies have addressed this form of feeding with at least 30 min of duration (Bueno et al. 2020; Ren et al. 2017a, b, c, 2018; Wei et al. 2012). As a result, the smallest phosphorus removals were found in the work of Di Bella and Torregrossa (2014) (Table 4).

Less explored but with great potential to favor granulation at high loads, staggered feeding is a strategy that can benefit conventional and autotrophic denitrification. It ensures greater stability to granules by reducing the influent load that can be toxic to microorganisms, distributing it throughout the cycle. Staggered feeding associated with SBR affects the selection and growth of filamentous organisms, playing a critical role in granule structure and composition (McSwain et al. 2004; Corsino et al. 2016). Generally, floc-forming bacteria with relatively high substrate absorption kinetics have an advantage over filamentous bacteria if the feeding is distributed. It forces the bacteria to acquire and store substrate for maintenance and cell growth during periods of famine, favoring the selection and formation of aerobic granules. In addition, by allowing nitrification to occur with a lower organic load in the aerobic phase, this feeding mode accelerates the nitrification rate and saves the aeration consumption from oxidizing the influent organic matter (Chen et al. 2011; Wang et al. 2012).

Anaerobic/anoxic phase and intermittent aeration

Regarding the quality of the granules formed, both the type of cycle and the distribution of phases throughout the cycle are preponderant factors in the granulation process. In addition to an entirely aerobic reaction phase, various operational configurations, such as anaerobic-anoxic-aerobic, anaerobic-aerobic-anoxic, and aerobic-anoxic conditions, have been adopted for wastewater treatment (Nancharaiah and Reddy 2018).

In the anaerobic/anoxic phase, nitrite and residual nitrate denitrification occur, in addition to EPS hydrolysis, fermentation, and VFA assimilation, with phosphate release. Being able to influence the granulation process, even in the anoxic phase, a low shear stress is imposed, different from what occurs in the aerobic phase when the granules are exposed to a greater shear stress caused by oxygen bubbles. Under aerobic conditions, a greater diversity of microorganisms starts to act in the granular biomass for simultaneous nitrification, denitrification, and phosphorus removal processes, such as AOB, NOB, common denitrifying heterotrophic microorganisms, PAOs, denitrifying PAOs (DPAOs), and denitrifying glycogen-accumulating organisms (DGAOs) (Nancharaiah and Reddy 2018; Rollemberg et al. 2018).

In order to favor DPAOs, which have a slower growth rate, the intercalation of anoxic and aerobic phases presents itself as a good strategy for high-strength effluents, and the exclusively anaerobic phase can be discarded since phosphorus concentrations are not high. Nancharaiah and Reddy (2018) suggest that integrating periods with high and low DO concentrations is necessary to achieve complete nitrogen removal. Zhang et al. (2014) point out that in anoxic phases, low phosphorus removal is due to the high presence of DGAOs, which compete directly with DPAOs.

In most studies using AGS to treat leachate, the anaerobic phase was non-existent, and when present, it was no longer than 90 min in long cycles (Table 5). However, there are no studies in which the anoxic phase has been used. Thus, the aerobic phase was an integral part of the reaction phase of AGS operating in SBR, and when operated in long cycles, it is expected that partial nitrification and complete denitrification can achieve higher rates, generating lower concentrations of toxic denitrification by-products and smaller accumulations of nitrate and nitrite.

Performance of AGS reactors in removing COD, nitrogen, and phosphorus from leachate

In AGS reactors, to simultaneously remove organic matter, nitrogen, and phosphorous species throughout the operational cycle, there must be a balanced oxygen supply to promote the nitrification without impacting the anoxic denitrification process or anaerobic phosphorus removal. Therefore, SBR with AGS can be adjusted to operate in cycle options: A/O (anoxic, oxic), A2/O (anaerobic, anoxic, oxic), or A/O/A (anaerobic, oxic, anoxic). The introduction of an anaerobic phase in the SBR operating cycle can reduce

Table 5 Efficiencies reported in the literature during leachate treatment with AGS technology

Influent (mg/L)				Operating parameters								Final removals (%)				NO_2^-acc.	References
COD	TN	NH_3-N	P_{Total}	Flow rate (L/min)	Reactor type	Cycle (h)	F (min)	AN (min)	AER (min)	S (min)	D (min)	COD	TN	NH_3-N	P_{Total}		
4298–5547	126–400	72–374	---	2	O	12	60	0	640	5	5	84	75	96	---	No	Wei et al. (2012)
4502–5992	691–1253	602–1168	---	2	O	12	60	0	640	5	5	83	35–58	39–77	---	Yes	
9738	3700	1960	---	---	O	24	5	0	1427	3	5	40–50	Low	40–60	---	Yes	Di Bella and Torregrossa (2014)
4560	1845	945	---	---	O	12	5	0	707	3	5	50–60	Low	20–50	---	Yes	
448–654	---	120–500	32.5	3	A2/O	8	30	90	345	5	12	66–73	39	61–95	34–54	No	Ren et al. (2017a)
448–654	---	120–500	32.5	3	A2/O	8	30	90	345	5	12	67–87	44–48	99	49	Yes	Ren et al. (2017b)
1080	---	340	2–6	2–3	O	8	90	0	360	5	12	65	40	100	80	Yes	Ren et al. (2017c)
1194	---	580	4–6	2–3	O	8	90	0	360	5	12	43	25	100	40	Yes	
1539	---	900	5–6	2–3	O	8	90	0	360	5	12	20	< 10	91	40	Yes	
550–1000	---	130–785	3–6	1.5	A2/O	8	30	90	330	5	12	43–65	24–37	95–99	0	Yes	Ren et al. (2018)
1000–1100	---	785–1085	3–6	1.5	A2/O	8	30	90	330	5	12	31–40	23–24	95–99	0	Yes	
1100–1200	---	1085–1209	3–6	1.5	A2/O	8	30	90	330	5	12	7–31	21–23	99	0	Yes	
650	114	88	13.1	1.5	O	8	45–60	0	425	5	5	87	99	99	36	No	Bueno et al. (2020)
863	187	136	15.2	1.5	O	8	45–60	0	425	5	5	89	99	99	42	No	
1421	304	281	17.5	1.5	O	8	45–60	0	425	5	5	88	98	99	45	No	

COD chemical oxygen demand, *TN* total nitrogen, *NH_3-N* ammoniacal nitrogen, P_{Total} total phosphorus, *F* feeding phase time, *AN* anoxic/anaerobic phase time, *AER* aerobic phase time, *S* settling phase time, *D* discharge phase time, NO_2^- *acc.* nitrite accumulation, *O* oxic, *A/O* anoxic, oxic, *A2/O* anaerobic, anoxic, oxic, *A/O/A* anaerobic, oxic, anoxic the aeration demand during the aerobic oxidation cycle (He et al. 2018).

After feeding, the reaction phase known as feast begins, i.e., the period when the external substrate is readily available and in abundance. This substrate diffuses inside the granule completely. Part of the carbon is converted and stored aerobically, anaerobically, or under anoxic conditions by heterotrophic microorganisms in the form of intracellular polymers such as PHB. Oxygen penetration into the granule is smaller than that of organic carbon due to the rapid consumption by autotrophic and heterotrophic organisms in the granule's outer layer. In this way, DO is used mainly for nitrification, aerobic carbon conversion, and biomass growth. The autotrophic organisms in the outer layers of the granules convert ammonia into nitrogen oxides (NOx) (nitrite and nitrate) that diffuse towards the granule center and into the liquid phase. In the anaerobic zone, PHB is available to be used as a carbon source for the denitrification process (Nancharaiah and Reddy 2018; Rollemberg et al. 2018). Therefore, nitrogen removal occurs through the distribution of phases and inside the granules via SND process.

The famine period starts after the complete consumption of organic matter. Oxygen penetration into the granule will be greater since its concentration in the liquid medium will be higher, and only PHB will be available. The oxidation of the stored PHB will occur through NOx production at a level that allows its introduction into the anaerobic zone, making it anoxic. This is due to high oxygen consumption through nitrifying autotrophic organisms. Thus, PAO and GAO will have access to oxygen, and DPAO and DGAO to NOx (Nancharaiah and Reddy 2018; Rollemberg et al. 2018).

Phosphorus removal occurs by the accumulation of polyphosphate in PAO and DPAO, requiring an anaerobic condition to favor PAO development and ensure that it prevails over GAO since both compete for substrate during the feast, and for DPAO to prevail about DGAO, as they compete for NOx during the famine (Nancharaiah and Reddy 2018).

Some recent studies have shown that COD and nitrogen removal efficiencies in AGS systems depend on several aspects, such as inoculum quality and cultivation strategy, system operation, and mixing/equalization of leachate with sanitary sewage (Bueno et al. 2020). The efficiency of the main AGS systems on leachate treatment is shown in Table 5.

In the first studies using AGS technology to treat landfill leachate, Wei et al. (2012) and Di Bella and Torregrossa (2014) showed that the process efficiency depends on the influent load since by increasing influent ammonia concentration, the removal rates of ammonia itself, total nitrogen, and COD tended to fall. Even so, the results indicated that AGS reactors easily achieved the removal of these compounds. However, it is important to note that leachate characteristics are key for system performance. For example, some studies have observed a low COD removal rate associated with a low leachate biodegradability, characteristic of old leachates (Di Bella and Torregrossa 2014; Wei et al. 2012).

Kocaturk and Erguder (2016) reported that the COD/ammonia ratio in the influent could influence the variety of dominant microorganisms in the granules, and the higher this ratio, the greater the COD removal. This was evidenced by Ren et al. (2017b), in which the COD removal efficiency decreased by 20% by reducing the COD/ammonia ratio in leachate from 5 to 1.5.

Yang et al. (2004) and Ren et al. (2017b) observed that free ammonia in the wastewater affects sludge and aerobic granules' aggregation and biomass washing by decreasing cellular hydrophobicity. In conventional biological systems (e.g., activated sludge), the inhibitory effects of free ammonia on the activity of nitrifying microorganisms were observed in concentrations above 10 mg/L for AOB and from 0.1 to 1.0 mg/L for NOB (Anthonisen et al. 1976; Yang et al. 2004; Zhou et al. 2011). However, in AGS reactors, it has been observed that the total ammonia removal efficiency remains high and stable, even in high ammonia concentrations. Such a characteristic is due to the compact and unique structure of the AGS granules, preventing nitrifying microorganisms from having direct contact with these toxic compounds (Ren et al. 2017c). In all cases, it is important to note that the low removal of TN can also be associated with the low availability of biodegradable COD, especially in old leachate, which decreases the carbon available to denitrifying microorganisms, thus hindering denitrification and SND (Ren et al. 2018).

Finally, Ren et al. (2017c) also found that phosphorus removal also decreases with the increase in leachate pollutants load. Muszyński and Miłobędzka (2015) point out that during high-load SBR cycles, it is expected that phosphorus removal will be opposite to nitrogen removal, possibly due to the competition for carbon between denitrifying heterotrophs, GAOs, and PAOs. The excess of nitrate in the anaerobic phase also causes this competition (Ren et al. 2018). In addition, it has been reported that the high presence of ammonia also inhibits PAOs, even at concentrations below 1 mg/L (Saito et al. 2004). Thus, low phosphorus removals have been observed (Bueno et al. 2020; Ren et al. 2017a, b, 2018).

Main challenges in the application of AGS in the treatment of leachate

Nitrite accumulation

In the SND process, the critical step is to obtain stable nitrification. Nitrite accumulation has been widely reported in experiments with high ammonia loads, decreasing total nitrogen removal. Nitritation is affected by the control of the reaction conditions (in a macro perspective) or by the rapid growth of AOBs to the detriment of NOBs (Miao et al. 2019; Poot et al. 2016). Studies show that NOBs have a slower growth rate than AOBs, the latter with more oxygen affinity (Ma et al. 2016; Regmi et al. 2014). Therefore, low concentrations of dissolved oxygen prevent NOBs from developing. According to Ren et al. (2016) and Miao et al. (2019), partial nitrification depends on pH, temperature, and free ammonia at higher levels, intensifying AOB activity.

Therefore, in leachate from old landfills, due to high concentrations of N-NH_3, partial nitrification (nitritation) has been the most effective route (Miao et al. 2014). Such a fact occurs since high levels of NH_3-N can increase the amount of free ammonia, generating a lot of NO_2-N, which allows the availability of high concentrations of free nitrous acid. According to Chung et al. (2015), all microorganisms responsible for nitrification and denitrification are affected by FA and FNA, with NOBs being the most susceptible. Some strategies have been reported, and by inhibiting free ammonia, Wu et al. (2015) managed to obtain a stable nitrification by treating municipal sewage and leachate simultaneously.

As mentioned, pH control can be the key factor for good nitrification, as it varies considerably during the process (Miao et al. 2019). When aeration begins, readily biodegradable organic matter is quickly degraded due to the high affinity of heterotrophic bacteria for oxygen. Subsequently to the COD consumption comes the conversion of ammonia to nitrite by autotrophic bacteria, in which the pH decreases as alkalinity is consumed. When this conversion ends, the pH rises again due to nitrite conversion into nitrate by the nitrite-oxidizing microorganisms, which do not require alkalinity consumption. Wang et al. (2013) point out that the pH is directly related to nitritation, conditioning the conversion of nitrite to nitrate. Thus, when adopting pH and DO control strategies, nitritation and denitritation efficiencies to over 90% were obtained (Chung et al. 2015; Sun et al. 2015; Chen et al. 2016).

Therefore, since AOBs occupy the surface of the granules and NOBs the interior, NH_3-N is immediately converted by AOBs (dominant group), and NOBs undergo two simultaneous inhibitions: by FA and FNA. In addition, the low DO concentration in the granule also influences NOB inhibition. All of this results in an accumulation of nitrite.

Biomass formation time

The high influent loads have significantly influenced the granulation process, especially organic matter and nitrogen. When working with landfill leachate, the required time to form a predominant granular biomass is relatively longer than in conventional effluents, such as domestic sewage. The granules' size is also smaller with uneven surfaces.

For example, Ren et al. (2018) and Bueno et al. (2020) needed 70 to 90 days of operation to obtain a granular biomass with characteristics similar to those of Yu et al. (2014) and Corsino et al. (2016) with less than 40 days of operation in influents with low concentrations of organic matter and nitrogen. Di Bella and Torregrossa (2014) and Bueno et al. (2020) observed the formation of more resistant and stable aggregations with diameters of 261–621 μm, only after 60 days of operation.

Generally, in the first 40 days of operation with leachate, the sludge has a predominant filamentous structure with a dark brown flocculating structure, low diameter, and minor microbial aggregations (Bueno et al. 2020; Ren et al. 2017a). Due to these characteristics of the biomass, solids washout tends to occur more frequently, delaying the granulation process (TSS loss greater than biomass growth).

Ren et al. (2017a) had so much sludge loss that it was impracticable to measure AGS reactors' sludge age. Bueno et al. (2020) chose to recirculate daily the sludge lost to the reactor to favor the granulation process, thus suggesting a secondary clarifier after the AGS reactor for automatic and efficient recirculation. Initial granulation using synthetic effluents with lower loads to generate stable and mature granules that can withstand hydraulic shocks before leachate incorporation into the AGS reactors is also suggested (Di Bella and Torregrossa 2014). Therefore, the treatment strategy must be adapted to minimize sludge loss and favor uniform granulation with large size granules.

Granule instability and disintegration problems

Recalcitrant wastewaters may cause inhibitory effects on AGS biomass, resulting in a low process yield (Li et al. 2014; Zheng et al. 2020). In wastewaters with high pollutant loads, especially landfill leachates, the main problems observed revolve around the disintegration and instability of granules. Several authors point out that these limitations are reflections of intracellular protein hydrolysis, as well as the destruction of EPS structure (PN proteins being essential for granules stability), anaerobic fermentation of dead and lysed cells in already stable granules, and degradation in the granule nucleus by the overgrowth of filamentous microorganisms at high rates of substrate transport, making them more floccular (Leal et al. 2020; Long et al. 2015; Lv et al. 2014; Wagner et al. 2015; Zhang et al. 2015).

It is important to note that the granular structure instability can compromise the treatment process efficiency since disintegration increases the TSS in the effluent and worsen the biomass settling in the mixed liquor (Li et al. 2014; Sarvajith et al. 2020). Furthermore, this instability can reduce the pollutant removal rate.

High organic loads accelerated the granulation process. However, this biomass outbreak generated rapid granules disintegration due to its excessive growth and subsequent increase of dead cells in the center and methanogens in the granule core (Long et al. 2015; Hamza et al. 2018).

In studies with leachate, Ren et al. (2017a) and Ren et al. (2018) found that granules formation occurred more slowly than the conventional one and that the granules formed were very unstable in load shocks. This profile was similar to other types of high-strength effluents. For example, Zhang et al. (2011) fed an AGS (SBR) with effluents from a petrochemical company and found that the granules' properties and performance declined with the shock.

Ren et al. (2017b) point out that in leachate, even after reaching operational stability, if there are significant changes in load, the granules disintegrate, and the denitrification process is compromised. Therefore, in effluents with high concentrations of organic matter or enriched with nitrogen/phosphorus species, instead of the available substrate being transformed by anaerobic processes into storage constituents, this substrate is adsorbed on the granules surface, favoring the development of filamentous structures in the granules and making them more susceptible to breakage (Pronk et al. 2015; Corsino et al. 2018). Therefore, granular disintegration remains an unsolved problem with AGS technology, and it is necessary to develop an efficient strategy for the direct treatment of these wastewaters with high resistance components. Hamza et al. (2018) propose selecting slow-growing organisms, such as phosphate-accumulating bacteria or glycogen, to maintain the granule stability upon high shock loads. In this regard, Wei et al. (2012) suggested that in leachate subjected to a physicochemical pre-treatment, the granules' disintegration was lower and the system achieved stability in time similar to the conventional low-load granulation.

López-Palau et al. (2012) found that an appropriate balance between the feast and famine phases must exist for the granules to be maintained in long-term operation. However, the duration of the substrate availability phase should not be greater than 25% of the total cycle period to guarantee the granule stability and an influent of adequate quality (Corsino et al. 2018). In addition to these operational adjustments, one of the most recently used strategies that have achieved excellent results is the formation of granules with gradual increases in loads and the dilution of real high-strength effluents before being subjected to treatment with AGS (Hamza et al. 2019; Leal et al. 2020; Ren et al. 2016; Xiong et al. 2020).

Conclusions and prospects for future work

The ability of aerobic granular sludge to withstand high carbon and nitrogen loads without significantly interfering in the removal efficiencies and biomass sedimentation shows that this technology has lots of potential and offers an excellent alternative to the conventional biological treatments for landfill leachates.

Nonetheless, there is still a significant knowledge gap regarding the optimal operating conditions to maximize the AGS-SBR reactor's performance in leachate treatment without high costs, i.e., minimizing energy consumption and the addition of reagents. The reactor configuration (conventional SBR versus simultaneous fill/draw SBR), feeding mode (continuous or intermittent), the influence of feeding concentrations (dilution rates), and cycle time (longer operation cycles, increment of anoxic phases, among others) are some of the aspects that need further exploration to employ AGS technology for landfill leachate treatment.

It is worth noting that the variability of leachate composition with the landfill age may require different treatment strategies. Nitrite and nitrate accumulation and total nitrogen removal can represent a problem for these systems, especially for old landfill leachates. Thus, future research must fully understand the AGS system behavior when treating both young and mature leachates, including the need for the following: (i) pre- and/or post-treatment, (ii) leachate dilution, and (iii) supplementation with external carbon/nutrients sources. Moreover, granules' formation and stability using high-strength wastewaters and/or leachate, rather than synthetic or domestic wastewater, must be evaluated.

Finally, it is extremely important to investigate the effect of such complex wastewater on the microbial populations involved and respective kinetics after long-term exposure.

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Vicente E.P.S.G. da Silva, Silvio L.S. Rollemberg, Sara G.S. Santos: writing — original draft ; writing — review and editing

Tânia F.C.V. Silva, Vítor J.P. Vilar, André B. dos Santos: writing — review and editing; funding acquisition

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Data availability All data generated or analyzed during this study are included in this article.

Declarations

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