The applications of aerobic granular sludge for leachate treatment: A review

To cite this article: D Ilmasari and A Yuzir 2022 *IOP Conf. Ser.: Earth Environ. Sci.* **1091** 012051

The applications of aerobic granular sludge for leachate treatment: A review

D Ilmasari[1], A Yuzir*[1],

[1] Malaysia-Japan International Institute of Technology (MJIIT), Universiti Teknologi Malaysia, Jalan Sultan Yahya Petra, 54100, Kuala Lumpur, Malaysia

*Corresponding author e-mail: muhdaliyuzir@utm.my

Abstract. The vast majority of the world's daily municipal solid waste (MSW) is dumped in landfills without being treated first. Leachate generated from these landfills is defined as dark-coloured, highly contaminated wastewater that has become a problem for the environment, posing a threat to soil, surface water, and groundwater quality and having a detrimental influence on human health. Thus, leachate treatment is strongly recommended prior to final discharge. The application of aerobic granular sludge (AGS) technology for wastewater treatment has increased in recent years, especially for industrial wastewater such as leachate. Due to its significant advantages over conventional activated sludge technology, such as denser structure, improved settleability, faster effluent separation, higher biomass content, and improved shock loading resistance, AGS is a viable option for leachate treatment. This article provides detailed discussions of the leachate characteristics along with the available treatment systems, focusing on the AGS system. The efficacy of AGS technology in the treatment of landfill leachate was elucidated by highlighting its benefits, governing factors affecting its performance, and limitations. Based on the latest literature, this paper attempts to identify the research gaps and obstacles in using AGS technology for landfill leachate treatment.

1. Introduction

Municipal solid waste (MSW) management is a severe environmental, social, and economic concern worldwide [1]. Most MSW is dumped in landfills, which are well-acknowledged for their simplicity and economic benefits [2]. However, the generation of leachate is one of several issues linked with landfills. Landfill leachate is a dark-coloured complex effluent generated from landfills that contains high levels of organic materials, ammonia, dissolved solids, heavy metals, and xenobiotic organic compounds [3]. It has been reported that leachate can cause a threat to soil, surface water, and groundwater quality and have a detrimental influence on human health [4]. Thus, leachate treatment is strongly recommended prior to final discharge.

Numerous methods are available for treating landfill leachate. One such approach is the biological treatment by activated sludge which has been extensively reported in the literature, achieving COD and ammonia removal up to 90% and 99%, respectively [5–8]. This approach, however, has been found to be inefficient as a stand-alone process, whereby nitrification is aided by activated sludge, while denitrification is reported to be less effective in reducing the nitrogen contents in leachate [9]. Disadvantages include excessive sludge production, high demand for operation cost, and microbial population inhibition due to leachate's high ammonia levels continue to be a severe challenge for its implementation [10].

Aerobic granular sludge (AGS) technology, which has been the subject of intensive research over the last decade, has been proven to be a novel and promising development in the field of wastewater treatment processes. Aerobic granulation is often accomplished in sequencing batch reactors (SBRs), with a cycle arrangement set to provide a rigorous selection for rapid settling sludge and frequent recurrence of feast and famine regimes resulting in the formation of dense and stable granules [11]. Due to the unique structure of AGS, including a distinct aerobic and anaerobic/anoxic layer within the granules, this system outperforms the conventional activated sludge, which has been confirmed by both lab-scale and full-scale studies [12]. Moreover, their excellent settling ability compared to sludge flocs is essential for successful sludge-effluent separation. AGS also has a higher and more stable metabolic rate, is resistant to shocks and toxins, and has long biomass residence periods [13]. In terms of treatment efficiency, AGS has shown a stable performance for BOD as well as nitrogen removal [14]. Furthermore, previous studies have reported high COD removal rate (>90%) from various types of wastewater, including leachate by AGS [15–17]. Therefore, AGS offers significant potential for landfill leachate treatment which its application has been studied in recent years. However, until the present, no comprehensive review of AGS utilization in leachate treatment has been reported.

This review presents detailed discussions of the leachate characteristics along with the available treatment systems, focusing on the AGS system. The efficacy of AGS technology in the treatment of landfill leachate was elucidated by highlighting its benefits, governing factors affecting its performance, and limitations. Based on the latest literature, this paper attempts to identify the research gaps and obstacles in using AGS technology for landfill leachate treatment, which shall substantially contribute to future studies of landfill leachate management.

2. Leachate production and characteristics

MSW is a general term for all waste generated by households [18]. The amount of MSW generated yearly on a worldwide scale is expected to hit 2.2 billion tonnes by 2025 [19]. Besides, the world's population is estimated to increase to 10 billion people in 2057 with an annual growth rate of 1.05%. This demographic increase adds significantly to the production of MSW, posing a serious environmental concern [20]. In terms of MSW management, landfilling, incineration, and composting are three of the most common disposal techniques [21]. Across many countries, particularly developing countries, sanitary landfills and open dump landfills are the most favored methods due to its economic benefits [22]. However, one of the most significant issues associated with landfills, in addition to numerous other drawbacks, is the production of toxic leachate [23].

Leachate is liquid that has seeped through waste piles that may have decomposed aerobically or anaerobically. The ensuing leachate is heavily polluted, possibly infiltrating into the ground and contaminating soil and groundwater. [24]. Some of the pollutants found in the leachate from landfills can be divided into four categories: dissolved organic compounds, inorganic macro-compounds, xenobiotic organic compounds, and heavy metals [25]. Leachate characteristics may vary from each site, whereby factors affecting their characteristics include landfill age, waste type and composition, seasonal variations, precipitation, and rainfall [26,27]. Several physicochemical parameters that are used to assess the quality of leachate such as biochemical oxygen demand (BOD), chemical oxygen demand (COD), total organic carbon (TOC) total suspended solid (TSS), total dissolved solid (TDS), ammonia (NH_3), total nitrogen (TN), total phosphorus (TP), chloride, sulfur compounds, alkalinity, and heavy metals [20–22]. Complex pollutants such as PFCs, PAHs, alkaline earth metals, and phthalic esters complicate the leachate composition even further [28].

Because leachate characteristics change over time, landfill age becomes crucial in leachate composition [29]. For example, BOD and COD concentrations continue to decrease with landfill age, possibly caused by organic waste decomposition in leachate [30]. Therefore, leachate is classified as young, intermediate, and mature based on landfill age [10]. As presented in Table 1, young acidogenic leachate is commonly characterized by a high level of COD (>10,000 mg/L) and has more easily biodegradable organic matter, indicated by BOD/COD ratio of >0.3. Meanwhile, mature leachate shows more stable leachate with a low concentration of biodegradable organic matter (BOD/COD ratio of <0.1). It is also observed that ammonia concentration does not decrease significantly over time and due to its toxicity, it may disturb the biological unit in leachate treatment, which indicate ammonia in leachate is a major environmental issue [31]. Because the quality and quantity of leachate varies, it is critical to implement dependable and effective treatment systems capable of dealing with such complex effluent before it can be safely discharged to the environment.

Table 1. Landfill leachate composition [3,32,33]

Parameters	Young (<5 years)	Intermediate (5-10 years)	Mature (>10 years)
COD (mg/L)	More than 10,000	4000-10,000	Less than 4000
NH_3 (mg/L)	Less than 400	-	More than 400
BOD/COD	More than 0.3	0.1-0.3	Less than 0.1
Total phosphorus (mg/L)	5-100	-	5-10
Heavy metals (mg/L)	More than 2.0	Less than 2.0	Less than 2.0
pH	Less than 6.5	6.5-7.5	More than 7.5
Biodegradability	Low to medium	Low	Low
Organic compound	80% VFA	VFA 5-30%, Humic acid, Fulvic acid	Humic acid and Fulvic acid

*VFA: Volatile Fatty Acids

3. Overview of landfill leachate treatment

Pollution of groundwater and surface water is a significant environmental issue arise from the discharge of landfill leachate [34,35]. The treatment of leachate is substantially more challenging than municipal wastewater because it contains humic compounds released during the decomposition of the organic matter of the waste [36]. Moreover, two main elements to consider while treating leachate are the volumetric flow rate and its composition [3]. Prior to releasing leachate into water bodies, it is critical to remove COD, BOD, and ammonium levels [25].

The treatment of leachate can be classified into two groups, including conventional and advanced treatment. Conventional treatment can further be divided into three sub-groups. First is the leachate transfer [37] as well as recycle or reinjection to landfill cell [38]. Second is the biological degradation processes which include aerobic and anaerobic process [39]. The third sub-group is the physicochemical processes such as adsorption, air stripping, coagulation/flocculation, advanced oxidation, chemical precipitation, etc [40]. Most conventional treatment approaches require multistage processes. Depending on the leachate characteristics, the treatment technique applied might be biological and/or chemical [3]. Meanwhile, leachate treatment utilizing membrane technology is considered as advanced treatment [41].

Biological degradation processes were reported to successfully remove 96% of organic matters [42] and 90% of inorganic content [43], respectively. Despite their ease of operation and low cost, their efficacy is limited to leachate with a high degree of biodegradability, which corresponds to young leachate [9]. Regarding physicochemical processes, chemical precipitation is one of the most applied processes that is easy to use and has a high reaction rate, usually faster than biological processes. This method, on the other hand, is often not effective at removing organic matters and total solids [44]. Occasionally, combining biological and physicochemical processes appears to be a viable option for reaping the benefits of both technologies.

4. Performance of AGS

The advantages of AGS have been highlighted as having a dense structure (Figure 1) with a high settling velocity, rich biomass retention, superior organics and ammonia removal, and resistance to shock loadings and hazardous substances [45]. AGS relies on microbial activity to remove organics, nitrogen, phosphorus, and other pollutants all at once in a single reactor (mostly in SBR), rather than different processes taking place in multiple treatment stages [46–48]. AGS does seem to be a more promising and energy-saving treatment alternative in the wastewater treatment industry, as well as for leachate treatment, given the occurrence of simultaneous nitrification denitrification and phosphorus removal (SNDPR) due to the presence of several redox conditions inside the granules (Figure 2) [17,49,50].

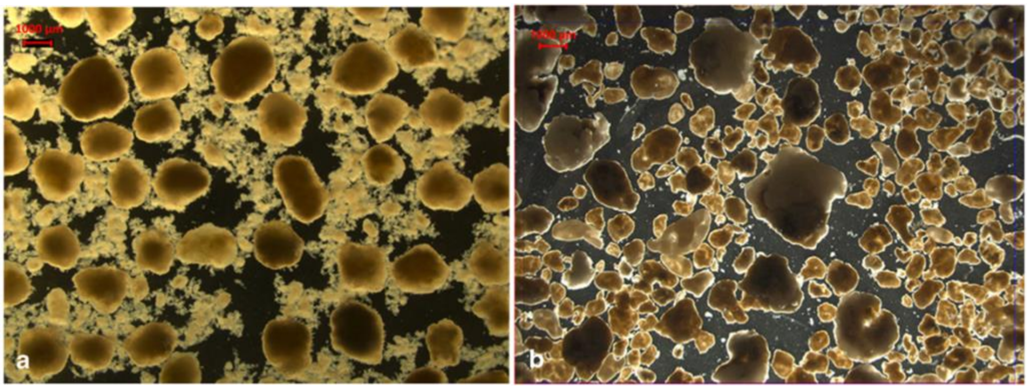

Figure 1. Stereoscopic view of AGS [51]

4.1. Performance of AGS for leachate treatment

For the treatment of leachate, AGS has demonstrated its effectiveness at removing organic matters and nutrients, e.g., NH_3 (Table 2). The system's ability to survive toxic loading allows it to be used for leachate containing a variety of contaminants. In general, the overall results of AGS treatment for landfill leachate are satisfactory. According to Ren et al. [52] AGS can remove 64% of COD, 95-100% of NH_3, and 84% of TP from leachate whereby the compact and distinctive shape of AGS may have aided in the almost complete NH_3 removal.

Wei at al. [53] conducted one of the earliest research on the treatment of leachate using AGS. The effect of varying NH_3 levels in influent on COD and nitrogen removal was investigated. By applying a modified kinetic model, COD removal efficiency was 76-91%, indicating that increasing NH_3 levels in the influent decreased the COD removal efficiency. Meanwhile, depending on the NH_3 levels in the influent, the nitrogen removal efficiency attained ranged from 40 to 92%.

Another study regarding the development of AGS for leachate treatment was conducted by Ren et al. [54]. By diluting leachate with municipal wastewater at a volumetric ratio ranging from 10 to 100%, leachate was gradually introduced to SBR. In comparison to the activated sludge system, the AGS system had a better settleability. Due to the refractory organic content in leachate, COD removal efficiency was low in both activated sludge and AGS. It was further revealed that AGS is more resilient than suspended-activated sludge in treating leachate with high NH_3 and free ammonia (FA) levels.

According to Wei et al. [55], the appearance of AGS seemed to increase the microbial diversity in the system. Some key and dominating populations in the activated sludge seed died out slowly, whereas a few microbial species emerged as the aerobic granules formed and matured. The newly emerged species were assumed to be reliable bacteria responsible for the high pollutant removal efficiency.

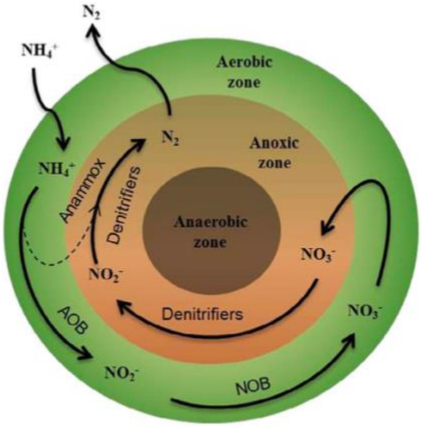

Figure 2. Nitrogen removal pathways inside AGS

Table 2. AGS for leachate treatment

Reactors	Leachate type (Influent feeding)	Removal efficiency	References
SBR	• Young leachate & synthetic wastewater (gradually added to SBR by dilution)	• COD:87-89% • NH_3:99% • TN:98% • TP:37-64%	[56]
SBR	• Old leachate & municipal wastewater • COD:1600-2850 mg/L • NH_3:1200 mg/L	• COD:31% • NH_3:99,7% • TN: 26%	[54]
SBR	• Synthetic young leachate • COD:498 mg/L • NH_3:120-500 mg/L	• COD:67-87% • NH_3:99% • TN:44% • TP:49%	[49]
SBR	• Synthetic old leachate & sewage (gradually added to SBR by dilution) • COD:1960-2850 mg/L • NH_3:704-1088 mg/L	• COD:64% • NH_3:95-100% • TN:42% • TP:84%	[52]
SBR	• Synthetic old leachate • sCOD:448-654 mg/L • NH_3:135-465 mg/L • TP:6 mg/L	• COD:55-73% • NH_3:61-95% • TN:39% • TP:54%	[50]
SBR	• Leachate diluted with tap water • COD:3996 mg/L • NH_3:1339 mg/L • TN:63%	• COD:84% • NH_3:89% • TN:63%	[55]
SBR	• Intermediate leachate • COD:4502-5992 mg/L • NH_3:257 mg/L (with pre-treatment) • NH_3:783-1079 mg/L (without pre-treatment)	• COD:82-84% • NH_3:92% (with pre-treatment) • NH_3:44-61% (without pre-treatment)	[53]
SBR	• Leachate diluted with tap water (1:1) and additional inorganic carbon added • COD:937 mg/L • BOD:589 mg/L • NH_3:303 mg/L	• COD:18-27% • TN:19-36% • NH_3:100%	[57]

4.2. Factors governing AGS treatment process for leachate

Several parameters, such as leachate toxicity, concentrations of organic matters and nutrients (i.e., NH_3) and leachate stability have been identified as the significant factors influencing AGS performance in leachate treatment process. The following sub-sections elaborate the factors governing AGS treatment efficiency.

4.2.1. Leachate age

Leachate aging causes the declining concentration of BOD; thus leachate becomes more stable due to the presence of refractory organic matter (e.g., fulvic and humid acid) as well as the decrease value of BOD/COD ratio [3]. Moreover, as landfills age, the non-biodegradable organic content of old leachate rises. As a result, biological treatment techniques may not treat old leachate as successfully as when it is applied for young leachate [30].

When AGS was used to treat old leachate, it was found that COD removal efficiency was as low as 31%. The findings indicated that the majority of COD in the old leachate was not biodegradable.

AGS, on the other hand, still outperformed activated sludge in terms of COD removal [54]. Similar findings were also obtained by Ren et al. [52] suggesting that low COD removal (64%) was associated with low BOD/COD ratios of old leachate varying from 0.15 to 0.2 (low biodegradability). On the other hand, when AGS was used to treat young and intermediate leachate, satisfactory COD removal was attained with 84% and 89% removal recorded (COD influent 4502-5992 mg/L) [53,56]. Based on these findings, AGS has the potential for old leachate treatment, yet there has been so no literature that study the improvement scheme to maintain the high performance of AGS in treating old leachate.

4.2.2. Ammonia nitrogen concentration

Under alkaline conditions, the existence of free ammonia (FA) is unavoidable due to the high concentration of ammonium in wastewater [58]. Meanwhile, various important bacteria essential for wastewater treatment are believed to be inhibited by the presence of FA [59]. Extracellular polymeric substances (EPS) which regarded responsible for the granules' chemical and physical properties, have been shown to be broken down by FA as well [60,61].

According to di Bella et al. [17], when ammonium concentrations in leachate rise to 2000 mg/L, organic and nitrogen removal tend to fall. Furthermore, a study by Wei et al. [55] revealed that ammonium removal from leachate by AGS was hindered when the high nitrogen loading rate (NLR) (1000-1500 mg/L) was applied, but ammonium removal was excellent at the low NLR (±500 mg/L). At the high NLR stage, the diversity and activity of microorganisms, particularly those of the NOB species, were suppressed. On the other hand, the microbial population stabilised at low NLR, ensuring efficient ammonium removal.

4.2.3. Temperature

An increase in temperature has been shown to increase wastewater treatment efficacy by aerobic granules [62,63]. Song et al. [64] reported that 30°C was the optimum operating temperature for better settleability, microbial activity, and granule compaction. The authors also imply that at various temperatures, different types of bacteria may play crucial roles. According to de Kreuk et al. [65], the growth of filamentous bacteria at 8-15°C causes granules to have an irregular structure, resulting in inadequate denitrification and other biodegradation processes. Furthermore, temperature influences the rivalry between phosphate accumulating organisms (PAO) and glycogen accumulating organisms (GAO) in granules [66].

Mieczkowski et al. [57] tested the effect of temperature on the removal of nitrogen and organic compound from landfill leachate. It was revealed that gradual adjustment of the granules to lower operational temperatures (29-20°C) allowed the granules to completely oxidise ammonium from leachate at ambient temperature (20°C), considerably improving denitrification efficiency. Aerobic and anaerobic ammonium-oxidizing bacteria both participated in ammonium oxidation. It was also revealed that the highest TN removal was achieved when the system was operated under 25°C condition. However, from all the temperature tested, COD removal was under 50%, indicating there is no significant effect of temperature on organic removal.

4.2.4. Leachate co-treatment

In the literature, the co-treatment of leachate and municipal wastewater using AGS has been discussed. This approach is being proposed as an alternative to overcome the shortcomings of biological leachate treatment [3]. Moreover, since it can be implemented at existing wastewater-treatment plants (WWTPs), this has been regarded as a practical and relatively low-cost option [3,67].

AGS treating young leachate that used the volumetric ratios of 5, 10, and 20% removed 89%, 99%, 98%, and 64% of COD, NH_3, TN, and TP, respectively. The COD and TN levels in the effluents increased as the leachate concentration increased from 5% to 20%. However, removal efficiencies were closely similar in all ratios tested, indicating that the increase in leachate had no significant effect on the treatment processes [56].

5. Limitations and future prospects

To this date, there is no treatment system that has been widely accepted and applied for leachate treatment. However, it is projected that AGS will likely overtake the conventional activated sludge system in the wastewater treatment industry. However, the implementation of AGS for industrial wastewater treatment, particularly for landfill leachate, still faces certain challenges.

- At present, most of AGS research has been done in lab-scale SBRs with defined substrates and well-controlled working conditions, utilizing synthetic wastewater. There are some studies that use actual leachate, but they are still deemed limited. Furthermore, the viability of adopting another type of reactor, such as continuous flow reactors, which are commonly used in WWTPs for leachate co-treatment, must be evaluated.
- Earlier research using AGS for leachate mainly focused on COD, nitrogen, and phosphate removal. The use of AGS to remove other pollutants such as heavy metals and phenolic compounds has yet to be investigated. Exploring the scopes of AGS application for leachate treatment is thus one of the next research directions.
- The operation parameters must be explored for varied leachate types. There are no standard operating criteria or specifications for AGS leachate treatment. Thus, future study should continue to focus on standardising treatment process.

6. Conclusion

AGS has good settling ability, low sludge generation, and high organic, nutrients, and toxic compound removal efficiency compared to conventional activated sludge. Furthermore, AGS has been widely utilized in the wastewater treatment industry and it is applicable for a wide range of wastewater types, including for industrial wastewater such as landfill leachate. However, the studies of AGS for landfill leachate treatment is still deemed limited. Future works should be conducted and focus on: (1) utilization of actual landfill leachate and various types of reactors, (2) exploring the application scope, (3) standardizing the operating parameters.

Acknowledgments

This work is funded by the AUN/SEED-Net Grant no. 4B403 under Collaborative Education Program for Sustainable Environmental Engineering Network (CEP-SEEN).

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