



Treatment of Landfill Leachate using Granular Multi-Stage Anaerobic Reactor: Optimisation through Response Surface Methodology

Aida Batrisyia Jasni^{1*}, Shreeshivadasan Chelliapan², Mohd Fadhil Md Din³ and Nithiya Arumugam²

¹School of Civil Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, Skudai, 81310, Johor, Malaysia

²Department of Engineering, Razak Faculty of Technology and Informatics, Universiti Teknologi Malaysia, Jalan Sultan Yahya Petra, 54100 Kuala Lumpur, Malaysia

³Centre for Environmental Sustainability and Water Security (IPASA), Research Institute for Sustainable Environment, Universiti Teknologi Malaysia, Skudai, 81310, Johor, Malaysia

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Abstract

One of the most hazardous sources of pollution these days is landfill leachate. This harmful wastewater is not only affecting the environment, but also the health of beings surrounding the landfills. Numerous treatments have been used to treat this recalcitrant wastewater; however, anaerobic treatment has been in focus in recent years. In this study, we investigated the interactive effects of chemical oxygen demand (COD), leachate percentage and pH on the performance of a granular multi-stage anaerobic reactor (GMSAR) treating landfill leachate. Response surface methodology (RSM) was utilised to project the interaction effects of the operating conditions of the treatment system in terms of COD removal and biogas yield. The optimum region of the GMSAR was acquired at influent COD of 1239 mg/L, a leachate percentage of 14.2% and a pH of 7.3. These variables resulted in a 71.9% COD removal and 65.9mL/d of biogas yield. The percentage of leachate and COD influent resulted respectively in the most effective parameters on the COD removal and biogas yield of GMSAR.

Keywords: Landfill leachate, Anaerobic treatment, Multi-stage anaerobic reactor, Response surface methodology, Biogas yield

1 Introduction

1.1 Landfill leachate

Most of the developing countries are facing a major problem in terms of waste disposal. The disposal of municipal solid waste (MSW) by dumping or burying it in a designated site is called landfilling. This, however, does not solve the problem as it basically transfers the MSW from urban areas to landfills (1). The disposal and improper management of MSW in landfills provide the opportunity for toxic substances in MSW to degrade into a liquid form called landfill leachate. Generally, landfills can be classified into five levels which are levels 0, I, II, III and IV. Table 1 shows the classifications of landfill sites in Malaysia. Out of 158 landfills nationwide, only 17 sites are categorised as sanitary landfills whereas the rest of the sites are classified as unsanitary landfills. Level II is regarded as a semi-sanitary landfill due to the absence of leachate treatment facilities (2). The first two levels of landfills are the most worrisome as the numbers of these type of landfills are the highest in the country. These landfills are today's main cause of groundwater pollution as they do not have any layer of protection to keep the leachate from seeping into the ground

(3). Leachate formed from water runoff at landfill often causes pollution to the soil, groundwater and surface water (4). Leachate enters the surroundings from the bottom of the landfill through unsaturated soil stratum and flows to the groundwater and eventually to surface water via hydraulic connections. The discharge of treated or raw leachate from the treatment facility may also taint the environment and affect public health (5).

Table 1: Classes of landfill sites in Malaysia (6, 7).

Level	Type of Landfill
0	Open Dumping
I	Controlled tipping
II	Sanitary landfill with bund and daily cover
III	Sanitary landfill with leachate recirculation system
IV	Sanitary landfill with leachate treatment facilities

Leachate is liquid rich in organic matter which can be distinguished by its colour, mainly brownish or viscous black. The quality of the leachate is influenced by several determinants such as age, precipitation, weather variation, waste type and

Corresponding author: Aida Batrisyia Jasni, School of Civil Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, Skudai, 81310, Johor, Malaysia; E-mail: aida@civil.my.

composition, depending on the lifestyle of the surrounding population and tip structure. Kurniawan *et al.* (8) mentioned that the composition of the landfill leachate depends greatly on the age of the leachate. As the harmful chemical substances found in leachate vary, they resulted in the increment of COD value, making them difficult to treat. The high COD value which not only contains organic matter but also inorganic matter requires a very intricate treatment system to treat the landfill leachate.

1.2 Stage anaerobic treatment

Various landfill leachate treatment methods have been studied such as physical, chemical and biological which consist of aerobic and anaerobic treatments. Recent years have shown a grown interest in the area of anaerobic treatments (9–11). Anaerobic digestion is a process carried out by microorganisms that are able to live in an oxygen-deprived environment. The disintegration of organic substance happens in four stages: hydrolysis, acidogenesis, acetogenesis, and methanogenesis.

In pursuance of optimising the environment in the reactor for the anaerobic bacteria and to boost the specific conversion reactions, physical separation is added in the sludge bed of a staged anaerobic treatment reactor. According to Lier *et al.* (12), studies have been conducted for the treatment of carbohydrate wastewater under mesophilic conditions by segregating the anaerobes involved in anaerobic digestion. The convenience of staging in this study is accredited to the high sludge yield of the carbohydrate fermenting bacteria. As the carbohydrate is fermented or pre-acidified in the first stage, a high volumetric fraction is passed on to the next stage for the methanogens to increase the methane yield of the anaerobic treatment system (12). The advantage of a staged anaerobic treatment system is also supported by Zhao *et al.* (13) who agreed that the anaerobic treatment is enhanced and needed only half of the amount of biomass in the conventional treatment system to double the activity of the anaerobes in the staged treatment system.

Intanoo *et al.* (14) mentioned that the arrangement of a stage anaerobic reactor provides an ideal surrounding for the breakdown of intermediates such as propionate, which is beneficial both in mesophilic and thermophilic treatment conditions. However, the type and sequence of stages should be thoroughly studied in the preliminary stage to select the optimum arrangement for a specific application. Nasr *et al.* (15) applied a staged process in an up-flow anaerobic sludge bed (UASB) reactor which resulted in a more stable thermophilic treatment system. Very low volatile fatty acids concentration and low hydrogen partial pressure were obtained in the last compartments. The simplest arrangement of a staged anaerobic reactor can be achieved by arranging two or more up-flow reactor in series. In recent years, an integrated staged reactor comprising of vertically and horizontally oriented reactor has been introduced in order to enhance the plug flow pattern and spatial biomass separation (16,17).

The efficiency of anaerobic treatment is further increased by providing the optimal conditions for wastewater treatment. Nonetheless, the optimisation process is conducted in an obsolete way where a parameter is changed while the others are kept constant, which is too resource-consuming (18). To overcome this problem, response surface methodology (RSM), a mathematical

and statistical tool, is applied for process modelling and optimisation studies (19). RSM has been extensively used in various studies as the design of experiment (DOE) and optimisation tools in anaerobic digestion (18–22). There is no information available on literature to the extent of the author's knowledge, on the treatment of landfill leachate using a stage granular anaerobic reactor. Therefore, this study used a unique four-stage granular multi-stage anaerobic reactor (GMSAR) for the treatment of landfill leachate, focusing on the COD removal and biogas yield. RSM was applied as a statistically based DOE to study the effects of COD influent, percentage of leachate and pH on the removal of COD and biogas yield. The optimal operating parameters for the COD removal and biogas yield were determined.

2 Materials and Methods

2.1 Wastewater characteristics

In terms of the feed substrate, this study used a mixture of meat and yeast extract for a start-up. In anaerobic treatment, the start-up of the reactor is an important phase for the stabilisation of the reactor. The substrate used for the reactor stabilisation is crucial in determining a successful start-up process. Various substrates have been used by researchers in order to smoothen and reduce the time taken for the start-up process. Most of the researchers used glucose as their substrate during the acclimatisation period. Though the glucose initially showed a good result in removing COD, the performance of the reactor deteriorated as the pH levels of glucose decreased abruptly (23–25). There are several researchers who used a different substrate for the start-up process such as meat extract (26). The characteristics of the meat extract are suitable for the start-up process as it contains vitamins, fats, carbohydrates, and protein. However, the COD removal was only 72% which was quite low (27). A study conducted by Zupancic *et al.* (28) revealed that yeast extract is a good co-substrate which enhances the biomethane production of wastewater. Therefore, this study used a mixture of meat and yeast extracts as substrates during the start-up and acclimatisation period. This combination of both meat and yeast extracts is theoretically able to increase the performance of the reactor during the start-up process in terms of COD removal and biogas yield. The feed used in this study is a matured landfill leachate supplied by Worldwide Landfill Sdn Bhd, Jeram, Selangor. The characteristics of the leachate are given in Table 2. The start-up of the GMSAR was accomplished using a mixture dilution of Bovril soup stock and yeast extract and the ingredients are shown in Table 3.

Table 2: Characteristics of matured leachate

	Results	Units
pH	8.0	-
Temperature	26.0	°C
COD	2500	mg L ⁻¹
BOD ₅ @ 20°C	486	mg L ⁻¹
Total Suspended Solids	220	mg L ⁻¹
Ammoniacal Nitrogen (AN)	717	mg L ⁻¹

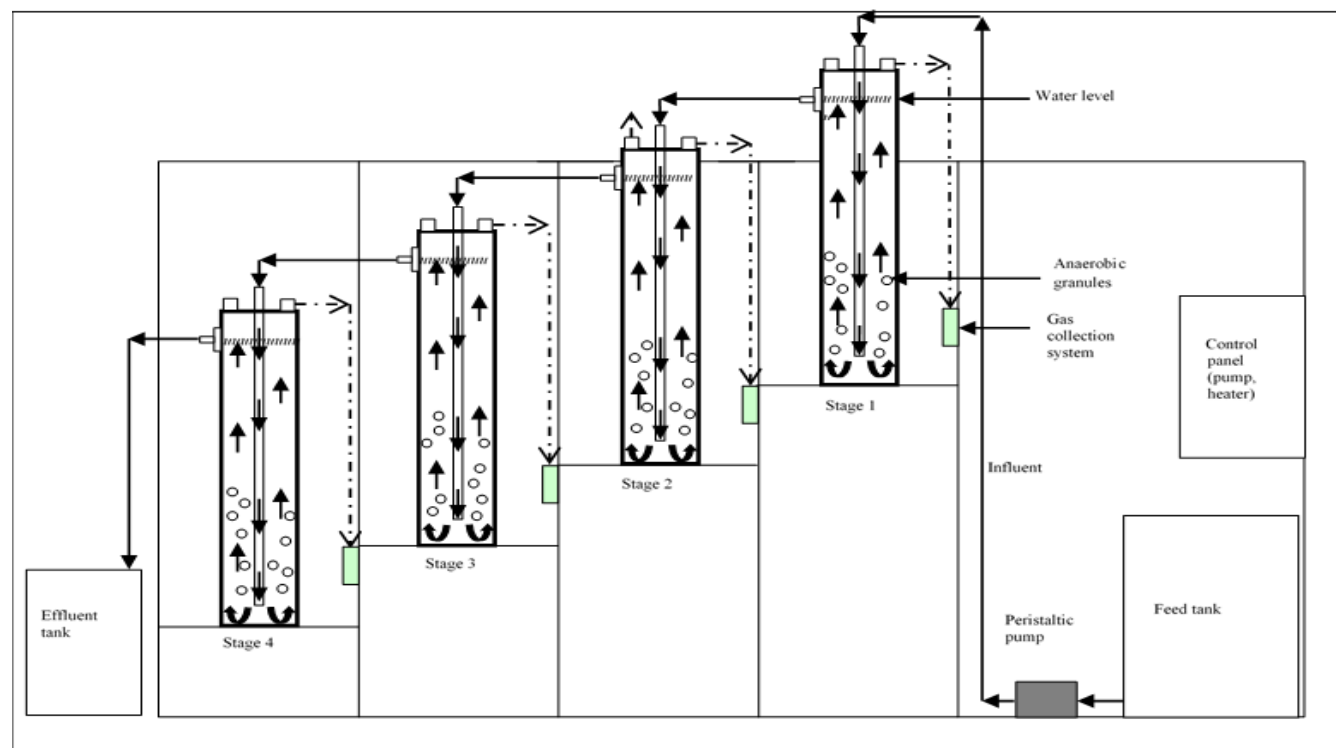


Figure 1: GMSAR configuration

Table 3: Ingredients of meat and yeast extract

Ingredients	Weight per 100 g
Yeast Extract	
Protein	38.4 g
Carbohydrates	19.2 g
Fat	0.1 g
Fiber	3.1 g
Sodium	4.3 g
Thiamine	5.8 mg
Riboflavin	7.0 mg
Niacin	160.0 mg
Folic acid	2500 µg
Vitamin B12	15.0 µg
Meat Extract	
Protein	13.3 g
Carbohydrates	24.4 g
Fat	0.1 g
Potassium	1200 mg
Sodium	3510 mg

Anaerobic granular sludge was used to seed the GMSAR. The utilisation of anaerobic granular sludge as the seed sludge in the present study was due to the efficient methane yield and pollutant removal as reported by Lu *et al.* (29). The anaerobic granular sludge contained a total suspended solid of 46,730 mg/L and 44,940 mg/L of total volatile suspended solids. The reactor was filled with tap water to the working volume. Subsequently, the reactor was flushed with nitrogen gas to displace residual air in the system before introducing the feed. The reactor was allowed to stabilise at 37°C for 24 hours for 7 days without further modification.

2.2 Reactor configuration

The GMSAR consisted of a working volume of 11.2 L and equipped with sampling ports at 15 cm from the base that allowed biological solid and liquid samples to be withdrawn from the sludge bed (Figure 1). The influent wastewater entered through a 2.7 cm internal diameter downcomer tube in the head plate that extended to within 55 cm of the reactor base and allowed feed to flow upward through the sludge bed. A temperature controller and heater strip were installed to retain the temperature at 37°C. Peristaltic pumps were used to control the influent feed rate to the GMSAR (Masterflex L/S, Easy Load II Pump Head). The GMSAR had anaerobic granular sludge as the sludge blanket.

2.3 Experimental procedure

The start-up of GMSAR lasted for 36 days. Initially, the reactor was inoculated with the granular sludge with 40% of the reactor's effective volume and then filled with tap water. After that, the GMSAR was left for about 8 days for the acclimatisation of the reactor. During the start-up of GMSAR, the feed was prepared using 30 ml of meat and yeast extract mixture and 18 L of tap water which gave a COD value of 300 mg/l. The pH profile of the reactor during the start-up showed stable performance at an average pH of 7.0, which confirmed that the reactor had a suitable pH level. As for the COD removal efficiency, the removal efficiency gradually increased to 90% and the leachate was gradually introduced into the feed for the treatment of landfill leachate.

The main objective of this study is to optimise selected variable factors including COD, percentage of leachate, and pH to evaluate the performance of the process by analysing COD removal and biogas yield as responses. As the variables concerned were only three, RSM was applied in the design of experiment using the

two-level full factorial design (FFD). In the two-level FFD, the number of experiments is 2^k , where k is the number of variables. Table 4 represents the value of factors in the experiment and the responses accordingly.

Table 4: The experimental plan of GMSAR and its raw responses result.

Run	Factors			Responses	
	COD Influent (mg/L)	Percentage of Leachate (%)	pH	COD Removal (%)	Biogas Yield (mL/d)
1	1239	0	7.3	89.7	59
2	270	30	6.6	5.12	11.2
3	1239	30	7.3	59	84.23
4	270	0	6.6	80.2	14
5	1239	30	6.6	23.5	37
6	1239	0	6.6	77.8	42
7	270	0	7.3	90.7	27.35
8	270	30	7.3	12.8	16.2

2.4 Analytical methods

To assess the efficiency of the GMSAR, the COD was measured based on the Standard Close Reflux Method using a HACH COD reactor described in the Standard Methods (No. 5220) (APHA, 2005) while the biogas yield was determined using an optical bubble counter (30).

3 Results and Discussions

3.1 Statistical analysis

The FFD method was chosen to attain the correlation between the manipulating parameters and the process responses. Table 4 shows the operating parameters involved (COD influent, percentage of leachate and pH) in terms of absolute units and the experimental data collected for the process responses (COD removal and biogas yield). In this study, eight experimental runs were performed by FFD in accordance with Table 4. The number of runs was determined using formulae 2^k , where k is the number of factors. The experimental results were then subjected to response surface analysis to investigate the interactive effects of parameters COD influent (A), percentage of leachate (B) and pH (C) on each factor. The regression model for the COD removal and biogas yield in coded terms, respectively, are as follows:

$$\text{COD Removal} = -0.001754A - 2.86536B + 23.42143C + 0.001169AB - 76.88524 \quad \text{Eq. (1)}$$

$$\text{Biogas yield} = 0.039598A + 29.49286C - 198.47919 \quad \text{Eq. (2)}$$

Eqs. (1) and (2) were figured out to attain the optimum value of each of the operating parameters to maximise the COD removal and biogas yield in the anaerobic treatment of landfill leachate using GMSAR. The adequacy and significance of the mathematical regression model were determined by analysis of variance (ANOVA). The results of the ANOVA are exhibited in Table 5. The F-value of the model for COD removal was 26.20, while the F-value for the biogas yield model was 14.38 which implied that both the models were significant. There were only 1.14% and 0.84% of chance, respectively, that the F-values this large could occur due to noise. The R^2 value of 0.9722 revealed that these particular mathematical models could explain 97.22%

and 85.19% of the variability in the COD removal and biogas yield responses respectively. Kainthola *et al.* (31) stated that the statistical model with R^2 value in the range of 0.75 – 1 shows that it is the best fit model. Adequate precisions in Table 5 measured the signal to noise ratio and a value of more than 4 was desirable. Adequate precision of the models was adequate as the values were 12.9235 and 8.3882 respectively. According to the model, the effect on each factor on the removal of COD was the percentage of leachate > pH > COD influent. On the other hand, the COD influent affected the biogas yield the most.

Table 5: ANOVA results for responses

Source	Sum of squares	df	Mean square	F-value	p-value
COD Removal					
Model	8662.44	4	2165.61	26.20	0.0114
A-COD Influent	467.87	1	467.87	5.66	0.0977
B-Percentage of Leachate	7079.31	1	7079.31	85.65	0.0027
C-pH	537.59	1	537.59	6.50	0.0839
AB	577.66	1	577.66	6.99	0.0774
R²	0.9722				
Adjusted R²	0.9351				
Predicted R²	0.8021				
Adeq Precision	12.9235				
Biogas Yield					
Model	3796.95	2	1898.47	14.38	0.0084
A-COD Influent	2944.51	1	2944.51	22.31	0.0052
C-pH	852.43	1	852.43	6.46	0.0518
R²	0.8519				
Adjusted R²	0.7927				
Predicted R²	0.6209				
Adeq Precision	8.3882				

3.2 COD removal and biogas yield

In this study of landfill leachate treatment using GMSAR, B, AB, C and A were ascertained as significant model terms for the COD removal. Percentage of leachate (B) had the highest effect on COD removal with an F-value of 7079.31 followed by the interaction between COD influent and percentage of leachate, pH, and COD influent with F-values of 577.46, 537.59 and 467.87, respectively.

The percentage of leachate played the biggest role in the removal of COD in this configuration of the anaerobic treatment system. As seen in Table 4, the COD removal decreased tremendously as the percentage of leachate injected into the stock solution increased. This result is corresponding to a study by Berenjkari *et al.* (32), whereby it was observed that the removal of COD decreased significantly with the increment of leachate into sewage sludge. The response surface plot for the relationship between the factors on the efficiency in removal of COD is shown in Figure 2. As can be seen, the COD removal decreases as the percentage of leachate increases. Though the COD influent was low, the high percentage of leachate in the stock solution reduced the removal of COD. This is consistent with the results of the variance analysis (Table 5). A high COD removal may have been

prevented by the toxicity of the landfill leachate itself which inhibited the acclimatisation of the microorganism to the recalcitrant wastewater (3).

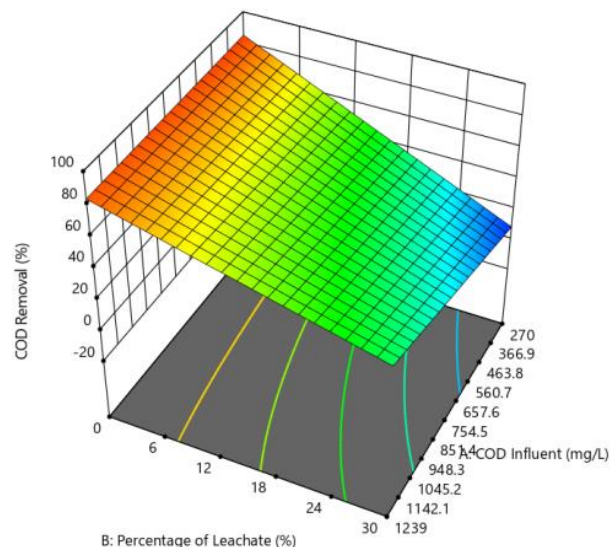


Figure 2: Surface plot of effects of COD influent and percentage of leachate on COD removal

Figure 3 displays the surface plot of the interaction of factors, COD influent and pH, on the biogas yield of this configuration of the landfill leachate treatment system. The most significant model term for the biogas yield was the COD influent which had an F-value of 2944.51, followed by pH with an F-value of 852.43. As discerned in Table 4 and variance analysis, the percentage of leachate did not affect the biogas yield. The largest contributor to the biogas yield was the COD influent. The higher the COD influent, the higher the biogas yield. The pH level also contributed to the amount of biogas yielded as the microorganism worked best in a near-neutral condition. The highest collection of the biogas yield in this study was recorded at 84 mL/d when the COD influent was 1239 mg/L and the pH was 7.3. The stages of GMSAR were for a better separation of acidogenesis and methanogenesis which theoretically, would be able to yield a high amount of biogas. However, the methanogens in the methanogenesis phase grew very slowly which reduced the consumption of volatile fatty acids (VFA), products from the acidogenesis stage. This resulted in the accumulation of VFA and inhibited the production of biogas (33, 34). This is similar to this study in which the biogas yield could be considered in the average region in comparison to the other types of anaerobic treatment system of landfill leachate (35,36).

3.3 Process optimisation

An experiment was conducted in the optimised condition as predicted by the model to validate its accuracy and the experimental data corresponded to the prediction of the model (Table 4). Under these optimal conditions (COD influent = 1239 mg/L, leachate percentage = 14.2% and pH = 7.3), the maximum COD removal and biogas yield were obtained as 71% and 65 mL/d, respectively. These results validated the accuracy of the model as the responses were comparable to the predictive values.

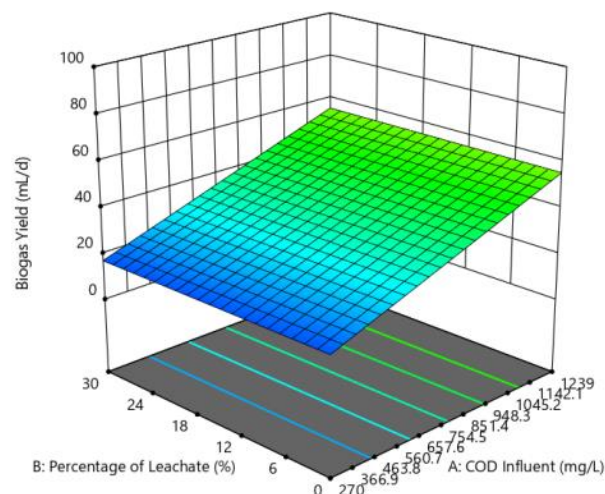


Figure 3: Surface plot of effects of COD influent and pH on the biogas yield

4 Conclusions

In this research, the GMSAR was found to be effective in biologically treating the landfill leachate. The RSM data showed the significant and interactive effects of the variables, COD influent, percentage of leachate and pH on the COD removal and biogas yield of the treatment. The results of the experiments revealed that GMSAR had the capability of treating the heavily polluted landfill leachate. The optimum conditions of the GMSAR were obtained at COD influent = 1239 mg/L, leachate percentage of 14.2% and pH at 7.3, at which the COD removal efficiency and biogas yield were 71% and 65 mL/d.

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Ethical issue

The authors are aware of and comply with the best practices in publication ethics specifically with regard to authorship (avoidance of guest authorship), dual submission, manipulation of figures, competing interests and compliance with policies on research ethics. The authors adhere to publication requirements that submitted work is original and has not been published elsewhere in any language.

Competing interests

The authors declare that there is no conflict of interest that would prejudice the impartiality of this scientific work.

Authors' contribution

All authors of this study have a complete contribution for data collection, data analyses, and manuscript writing

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