

# Performance of an Up-Flow Anaerobic Sludge Bed (UASB) Reactor Treating Landfill Leachate Containing Heavy Metals and Formaldehyde

Sivathass Bannir Selvam<sup>\*</sup>, Shreeshivadasan Chelliapan<sup>\*</sup>, Mohd. Fadhil Md. Din<sup>\*\*</sup>, Rafidah Shahperi<sup>\*\*</sup>, Maizatul Asnie Md. Aris<sup>\*</sup>

<sup>\*</sup>UTM Razak School of Engineering and Advanced Technology, Universiti Teknologi Malaysia, Jalan Semarak, 54100, Kuala Lumpur, Malaysia

<sup>\*\*</sup>Department of Environmental Engineering, Faculty of Civil Engineering, Universiti Teknologi Malaysia, 81310, Johor Bahru, Malaysia

Corresponding authors: [shreeshivadasan.kl@utm.my](mailto:shreeshivadasan.kl@utm.my) & [p56050@gmail.com](mailto:p56050@gmail.com)

## Abstract

Municipal Solid Waste (MSW) landfill leachate is a serious environmental issue and treated using various methods, mostly involving biological treatment. In the present study, an up-flow anaerobic sludge bed (UASB) was used for the treatment of matured landfill leachate that contains heavy metals (As, Fe, Ni, and Cd) and Formaldehyde (FA). Accordingly, the OLR to the UASB reactor was gradually increased from 0.125 to 2.5 kg CODm<sup>-3</sup>d<sup>-1</sup>, to observe the process performance. The process performance of the reactor was characterized in terms of pH, Chemical Oxygen Demand (COD) removal, Total Volatile Acid (TVA) production, Mixed Liquor Suspended Solids (MLSS), Mixed Liquor Volatile Suspended Solids (MLVSS) washout, and Methane composition. Results showed that at a Hydraulic Retention Time (HRT) of 4 days and an OLR of 0.125 kg CODm<sup>-3</sup>d<sup>-1</sup>, up to 79.04% COD removal efficiency was observed. However, when the OLR was increased gradually from 0.375 to 2.5 kg CODm<sup>-3</sup>d<sup>-1</sup>, the COD removal efficiency decreased to 9.5%, suggesting that the high accumulation of heavy metals may have inhibited the methanogens. During this period, the heavy metal and formaldehyde concentration were 9.40, 0.43, 0.50, 12.80 and 8.60 mgL<sup>-1</sup> respectively.

## Keywords

Heavy metals, formaldehyde, UASB, anerobic treatment, leachate, reactor performance

## INTRODUCTION

Landfills are important infrastructure along urban development, and the generation of leachates is an unavoidable condition. The actual landfill function is to store and manage solid waste effectively. The operations involving landfill leachate has grown to be a threat to the natural environment (Malyuba *et al.*, 2013). The leachate made by the rain along with exterior normal water movement in the landfill, through the time interval, and percolated and collected at the bottom of the landfill. The leachate can be a probable hazard to the good quality of groundwater and may pollute the groundwater by its contaminants. Landfill leachate can also be toxic to aquatic life.

Leachates are viscous black or brownish liquid rich in organic matter. The landfill leachate contains many chemicals, for instance, excessive phosphate, nitrates along with other metal salts. It also contains heavy metals such as As, Fe, Ni, and Cd, that can cause toxicity to the environment, flora and fauna (Malyuba *et al.*, 2013). A typical leachate contains a Chemical Oxygen Demand (COD) of 4,000

– 20,000 mgL<sup>-1</sup> depending on the maturity of the leachate. The permissible level of COD in wastewater as per Standard A and B are from 50-100 mgL<sup>-1</sup> (Environmental Quality Regulations-Industrial Effluents, 2009). There is a mixture of a chemical substance in leachate; thus a combination of treatment method is required. At the moment, leachate is treated by biological processes such as aerobic ponds or activated sludge reactors. However, the remaining values of COD and AOX are still relatively high. As a result, an alternative treatment method is preferred, and anaerobic treatment technology has emerged as one of the options. It has been used for over a century in the treatment of domestic and industrial wastewaters.

Anaerobic treatment is more preferable because it is the most suitable treatment option for high strength wastewater with comparatively less energy consumption and sludge production in addition to producing saleable end products like biogas and facilitating energy production. It is a biological process in which organic matter is degraded by a series of gaseous products, mainly CH<sub>4</sub>, CO<sub>2</sub> and H<sub>2</sub>, and its liquid effluent containing the most refractive compounds with significant presence of nitrogen, phosphorus and mineral compounds such as K, Ca and Mg. The conversion of the organic compounds to methane is a very complex process and requires the presence of different microbial species. Among the anaerobic reactors the up-flow anaerobic sludge bed (UASB) has been acknowledged as an alternative cost-effective process for the treatment of sanitary wastewater and wide varieties of high-strength industrial wastewaters. It is a robust technology and by far the most widely used high-rate anaerobic process for wastewater treatment.

The aim of this study was to treat the heavy metals and formaldehyde containing matured leachate by a UASB reactor. Specifically, the effect of increasing OLR to the reactor performance was evaluated in terms COD removal, effect on pH and volatile fatty acids, and methane composition. In addition, the heavy metals and formaldehyde concentration in the feed of the reactor system was measured during each OLR study.

## **MATERIALS AND METHODS**

### **Up-Flow Anaerobic Stage Reactor (UASB)**

The UASB used in this experimental study consists of 18 cm internal diameter by 110 cm height, with active volume of 20 L. The reactor had a 3-phase separator baffle, (2 circle disks with pore size diameter of 2 mm) and placed 2 cm below the effluent ports, to prevent floating granules from being washed out with the effluent. Sampling ports are placed at 8 cm intervals (lowest being 21 cm from the base) that allowed biological solids and liquid samples to be withdrawn from the sludge bed. The influent wastewater entered through a 2.7 cm internal diameter down comer tube in the head plate that extended to within 105 cm of the reactor base and allowed feed to flow upward through the sludge bed. Temperature controller and heater were installed to maintain the reactor temperature at 37°C. The UASB reactor can work in a wide range of temperatures, from mesophilic to thermophilic. For this study, mesophilic temperature 37°C was chosen to treat the matured leachate samples due to its treatment efficiency and lesser energy requirement.

### **Matured Landfill Leachate**

The matured leachate was obtained from an ageing leachate treatment pond in Jinjang transfer station, Selayang, Selangor and had the following characteristics: pH=8.0, COD=2500mgL<sup>-1</sup>,

Arsenic=9.40mgL<sup>-1</sup>, Iron=12.8mgL<sup>-1</sup>, Nickel=0.50mgL<sup>-1</sup>, Cadmium=0.43mgL<sup>-1</sup> and Formaldehyde=8.6mgL<sup>-1</sup>.

### Reactor Operations

The reactor was seeded with anaerobic digested sewage sludge from Bunus Sewage Treatment Plant. A total of 7 L of sieved sludge was added to the reactor after that the reactor was filled with tap water up to 20 L mark. Then the reactor was flushed with excess nitrogen gas to remove remaining air within the reactor space. The reactor was allowed to stabilize at temperature of 37°C for 24 hours. The start-up of the reactor was carried out using dilute leachate with very low COD concentration. Once the reactor attained a steady state condition (>80% COD removal), the feed (leachate) concentration was increased gradually by reducing the amount of water. The OLR was increased stepwise from 0.125 to 0.833 kg CODm<sup>-3</sup>d<sup>-1</sup> and increased further to 2.5 kg CODm<sup>-3</sup>d<sup>-1</sup> by reducing the HRT from 4 to 1 d. Finally, the OLR was reduced again to 0.375 kg CODm<sup>-3</sup>d<sup>-1</sup> (HRT 4 d) to determine the ability of the reactor to recover treatment efficiency. The optimum macronutrient to COD ratio was maintained at (COD: N: P, 250: 7: 1) by adding N100 macronutrient supplement and trace elements were not added due to natural presence of trace elements in the leachate.

### Sampling and Analysis

Sample analysis such as COD, pH, suspended solids (SS), and volatile suspended solids (VSS), were conducted according to Standard Methods (APHA, 1985). The total biogas volume was determined using an optical gas bubble counter. The biogas composition was determined using portable gas analyzer GA2000, Geotechnical Instruments. The FA content was analyzed using a HPLC with the following conditions: Column = Zorbax, C18, 4.6 mm x 250 mm ID, 5 µm particle size; Mobile phase = 70/30 acetonitrile/water (v/v); Flow rate = 1.2 mLmin<sup>-1</sup>; Detector = Ultraviolet, operated at 360 nm; and Injection Volume = 20 µL. The heavy metal analysis of the leachate was conducted by using an AAS according to EPA manual SW-846. The characterizations of the leachate samples were performed using High-Performance Liquid Chromatography (HPLC) 1220 Infinity LC by Agilent Technologies and Atomic Absorption Spectrometry (AAS) AA-7700 by Shimadzu.

Average values of the measured parameters quoted for each OLR were based on the mean of three data points taken when the reactor approached steady state.

## RESULT AND DISCUSSION

pH is an important parameter in an anaerobic treatment performance, and many studies have shown the optimum pH for the anaerobic digestion is in the alkaline region (Kawai et al., 2012). In addition, many anaerobic reactors could not tolerate acidic conditions due to the sensitivities of the methanogens. It is also well documented that the cause of pH drop in anaerobic reactors is due to the production of acidogenic bacteria. However, Rodriguez et al. (2011) demonstrated that methanogenic bacteria were able to produce bicarbonates to buffer the pH change caused by acidogenic bacteria. These hypotheses were compared in the present investigation. Figure 1 illustrates the pH variations and the effect of volatile fatty acids in the UASB when the OLR was gradually increased from 0.125 to 2.50 kg CODm<sup>-3</sup>d<sup>-1</sup>. The pH levels were generally stable (pH 8.17 – 7.52) in the UASB until the reactor OLR exceeded 0.625 kg CODm<sup>-3</sup>d<sup>-1</sup>. Consequently, at a reactor OLR of 0.833, the pH in the reactor dropped to 6.77 due to the rapid production of VFAs resulting from increased acidogenic activity. Further increase in the OLR from 1.25 to 2.50 kg CODm<sup>-3</sup>d<sup>-1</sup> deteriorated the pH of the reactor to 6.09 to 5.49. However, when the reactor OLR was reduced back to 0.375 kg CODm<sup>-3</sup>d<sup>-1</sup>, the pH in the reactor stabilised at 8.38

indicating that acidogenesis and methanogenesis had recovered balanced levels. From the pH data, it can be assumed that the metabolic processes differed between each OLR of the UASB system and this would cause each OLR to favour a unique population of microorganisms.

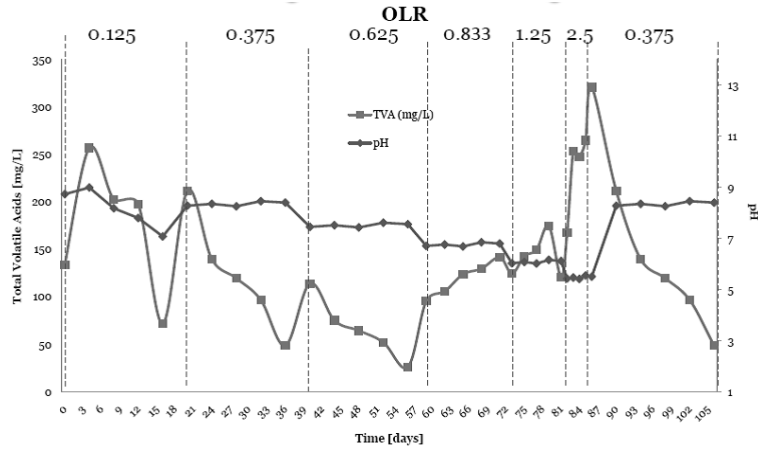


Figure 1. Variation in pH and Total Volatile Acids Performance

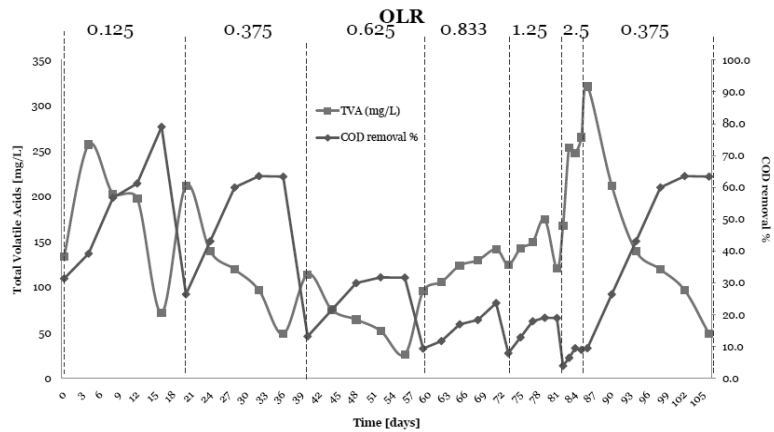


Figure 2. Variation in Total Volatile Acids and COD Removal Performance

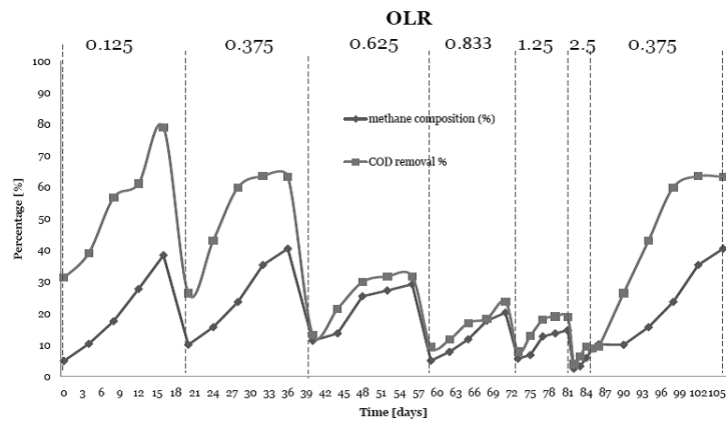


Figure 3. Comparison Methane Production and COD Removal Performance

At OLR 0.125 kg COD m<sup>-3</sup> d<sup>-1</sup> (HRT 4 d) the maximum overall COD removal efficiency was 79.04% (Figure 2). This condition considered acceptable during the acclimatization period of anaerobic digestion (Buitrón *et al.*, 2003; Enright *et al.*, 2005; Al-Karimiah *et al.*, 2011). Similar trend was also observed for the VFA profiles. Volatile fatty acid (VFA) concentration is an indicator of feed utilization by anaerobic microorganism (Deng *et al.*, 2013). When there is a build-up of VFA in the anaerobic system, probably it is an indication of anaerobic microorganism's failure to utilize the VFA as feed (Deng *et al.*, 2013). At initial stages of the OLR (0.125 and 0.375 kg CODm<sup>-3</sup>d<sup>-1</sup>) the VFA concentration was very high (200 mgL<sup>-1</sup>), however, this phase was temporary as the VFA utilization increased at the end of the respective OLR cycles. This could be due to the sudden shock to the microorganisms acclimating to the new type of feed substrate (leachate) as observed by Chelliapan *et al.*, (2006) during the treatment of pharmaceutical wastewater in an anaerobic reactor.

Further increase of the OLR from 0.375 to 1.250 kg COD m<sup>-3</sup> d<sup>-1</sup>, resulted in a low COD removal efficiency, until 9.5% was observed at an OLR of 2.50 kg COD m<sup>-3</sup> d<sup>-1</sup>. It is unlikely that this was caused by limitations in the UASB reactor as this reactor have been shown to achieve over 90% COD removal at high OLR (e.g. more than 20 kg COD m<sup>-3</sup> d<sup>-1</sup>). However, matured landfill leachate containing a high proportion of recalcitrant and complex organic carbon content, probably limits the UASB performance at high OLR. Moreover, heavy metals and FA concentrations in the feed (leachate) may have also contributed to the poor performance of the reactor system (Abbassi *et al.*, 2012; Mahmoudkhani *et al.*, 2011).

The maximum biogas composition (40.6%) observed at an OLR of 0.375 kg CODm<sup>-3</sup>d<sup>-1</sup> (Figure 3), probably due to the changes in the OLR, since the methanogenic bacteria is sensitive to changes in the feed OLR. As the indirect measure of biomass fluctuations in the reactor, the suspended solids in reactor correlate well with the methane generation (Raposo *et al.*, 2012). Apart from that the overall trend of the methane generation also matches the overall COD removal profile. Besides that, it was also observed that at all the OLRs, when the VFA concentration increased, the methane generation also decreased. However, when the VFA concentration decreased the methane concentration increased (Guendouz *et al.*, 2012). The methane profile has a close relationship with pH where a decrease in the pH will affect the methane generation (Xu *et al.*, 2011).

Table 1: Effect of inhibitor concentration on COD removal, pH, VFA and Methane generation

OLR (kg CODm <sup>-3</sup> d <sup>-1</sup> )	Feed concentration (mgL <sup>-1</sup> )					Max. COD Removal (%)	Max. CH <sub>4</sub> (%)	Mean VFA (mgL <sup>-1</sup> )	Mean pH
	As	Cd	Ni	Fe	FA				
<b>0.125</b>	0.47	0.0215	0.025	0.64	0.43	79.04	38.50	200.0	8.17
<b>0.375</b>	1.92	0.0645	0.075	1.92	1.29	63.50	40.60	91.67	8.36
<b>0.625</b>	2.35	0.1075	0.125	3.20	2.15	31.80	29.40	45.00	7.52
<b>0.833</b>	3.13	0.143	0.167	4.26	2.86	23.70	20.30	126.67	6.77
<b>1.250</b>	4.70	0.215	0.25	6.40	4.30	19.10	14.70	155.00	6.09
<b>2.500</b>	9.40	0.43	0.50	12.80	8.60	9.50	9.53	250.00	5.49
<b>0.375</b>	1.92	0.0645	0.075	1.92	1.29	64.10	39.98	90.00	8.38

Table 1 illustrates that the optimum conditions for the anaerobic digestion were at an OLR of 0.125 kg CODm<sup>-3</sup>d<sup>-1</sup>. The methane production was optimum at an OLR of 0.375 kg CODm<sup>-3</sup>d<sup>-1</sup>. Microorganisms were known to have the ability to bind with metals including toxic heavy metals (Abdel-Raouf *et al.*, 2012). A study by Lu and Hegemann (1996) demonstrated the inhibition of FA on anaerobic bacteria's at a concentration of 200 mgL<sup>-1</sup>. In a separate study carried out by Vidal *et al.*, (1999) the toxicity level of FA was determined as 100 mgL<sup>-1</sup>. As a result, at the current feed concentration of the FA (8.6mgL<sup>-1</sup>), it was thought that this concentration level in the reactor could be high at OLR of 2.50 kg CODm<sup>-3</sup>d<sup>-1</sup> (actual FA concentration in the UASB not measured), and may have contributed to the poor performance of the UASB reactor.

## CONCLUSIONS

This study demonstrates that the UASB reactor can be used to treat the matured landfill leachate. However, the treatment efficiency of the reactor was affected at an OLR of 2.5 kg CODm<sup>-3</sup>d<sup>-1</sup>, probably due to the recalcitrant nature of the wastewater that contains high heavy metals and formaldehyde at elevated OLR. At high OLR, the concentration of heavy metals and formaldehyde may have increased in many folds and may have inhibited the methanogens. Further studies should be carried out on the heavy metals and formaldehyde concentrations in the reactor as well as in the effluent.

## ACKNOWLEDGEMENTS

The authors desired to show the highest gratitude to Mr Azmi who helped with the apparatus and materials for the experiments and also, special acknowledgement to Universiti Teknologi Malaysia for funding this study. The authors also thank the Ministry of Education Malaysia for the Research University Grant (Vote number: 06H26).

## REFERENCES

- Abbasi, T., Tauseef, S.M., Abbasi, S.A., 2012. Anaerobic digestion for global warming control and energy generation—An overview. *Renewable and Sustainable Energy Reviews* 16 (9) 3228–3242
- Alkarimiah, R., Mahat, S.B., Md. Din, F., Chelliapan, S., 2011. Performance of an innovative multi-stage anaerobic reactor during start-up period. *African Journal of Biotechnology* 10 (54) pp. 11294-11302.
- American Public Health Association, 1985. In: Greenberg, A.E., Trussell, R.R., Clisceri, L.S. (Eds.), *Standard Methods for Examination of Water and Wastewater*, 16th ed. Washington, DC.
- Chelliapan, S., Wilby, T., Sallis, P.J., 2006. Performance of an up-flow anaerobic stage reactor (UASR) in the treatment of pharmaceutical wastewater containing macrolide antibiotics. *Water Research* 40 (8) 507 – 516
- Correa, S.M.B., Ruiz, E., Romero, F., 2003. Evolution of operational parameters in a UASB wastewater plant. *Water SA* 29 (4) 345-352
- Deng, Y., and Ezyske C.M., 2013. Sulfate radical-advanced oxidation process (SR-AOP) for simultaneous removal of refractory organic contaminants and ammonia in landfill leachate. *Water Research* 45 (3) 6189-6194
- Environmental Quality (Industrial Effluents) Regulations, 2009 Fifth Schedule [Paragraph 11(1) (A)] Acceptable Conditions for Discharge of Industrial Effluent for Mixed Effluent of Standards A

and B

- Guendouz, A.A., Brockmann, D., Trably, E., Dumas, C., Delgenes, J.P.S., Escudie, R., 2012. Total solids content drives high solid anaerobic digestion via mass transfer limitation. *Bioresource Technology* 111 (4) 55–61
- Kawai, M., Kisshi, M., Hamersley, M.R., Nagao, N., Hermana, J., Todaa, T., 2012. Biodegradability and methane productivity during anaerobic co-digestion of refractory leachate. *International Biodeterioration & Biodegradation* 72 (4) 46-51
- Lu, Z., and Hegemann, W., 1998. Anaerobic Toxicity and Biodegradation of Formaldehyde in Batch Cultures. *Wat.Res.* 32 (4) 209-215
- Mahmoudkhani, R., Hassani, A.H., Borghei, S.M., 2011. Study on Anaerobic Landfill Leachate Treatability by Membrane Bioreactor. *International Conference on Biology, Environment and Chemistry* 1 (3) 5-9
- Malyuba, A.D., Hani, A.Q., Hatem A., 2013. Assessment of Heavy Metals and Organics in Municipal Solid Waste Leachates from Landfills with Different Ages in Jordan . *Journal of Environmental Protection*, 4, (2) 344-352
- Raposo, F., De la Rubia, M.A., Borja, F.C.R., 2012. Anaerobic digestion of solid organic substrates in batch mode: An overview relating to methane yields and experimental procedures. *Renewable and Sustainable Energy Reviews* 16 (3) 861–877
- Rodriguez, R., Moreno, L., 2011. Simulation of a UASB reactor. *Tenth International Conference on Modelling, Monitoring and Management of Water Pollution*, Bucharest, WIT Press, pp 301-310.
- Vidal, G., Jiang, Z.P., Omil, F., Thalasso, F., Mendez, R., Lema. J.M., 1999. Continuous anaerobic treatment of wastewaters containing formaldehyde and urea. *Bioresource Technology* 70 (8) 283-291
- Xu, S.Y., Lam, H.P., Karthikeyan, P., Wong, J.W.C., Optimization of food waste hydrolysis in leach bed coupled with methanogenic reactor: Effect of pH and bulking agent. *Bioresource Technology* 102 (6) 3702–3708