Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2017.21406

86 (2017) 51–58
August

Performance of an up-flow anaerobic sludge bed (UASB) reactor for treating landfill leachate containing heavy metals and formaldehyde

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Received 2 September 2015; Accepted 2 July 2017

ABSTRACT

Municipal solid waste landfill leachate is of serious environmental concern and is 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). The organic loading rate (OLR) to the UASB, as measured by the chemical oxygen demand (COD), was gradually increased from 0.125 to 2.5 kg m^{-3} d^{-1}. The process performance of the reactor was characterized in terms of pH, COD removal, volatile fatty acid (VFA) production and methane composition. Results showed that with a hydraulic retention time of 4 d and an OLR of 0.125 kg COD m^{-3} d^{-1}, up to 79.04% COD removal efficiency was observed. However, when the OLR was increased gradually from 0.375 to 2.5 kg COD m^{-3} d^{-1}, the COD removal efficiency decreased to 9.33%, suggesting that the accumulation of heavy metals may have inhibited the methanogenic microorganism activity. Under the high COD loading conditions, the heavy metal and FA concentrations were 9.40 (As), 0.43 (Fe), 0.50 (Ni), 12.80 (Cd) and 8.60 (FA) mg L^{-1}. The removal of Cd, Ni and Fe was almost constant regardless of the OLR (around 36% for Cd, 32% for Ni and 29% for Fe). As and FA displayed a degree of removal at low OLR (40% and 17% at 0.125 and 0.833 kg COD m^{-3} d^{-1}, respectively), but at high OLR (2.5 kg COD m^{-3} d^{-1}), both As and FA decreased dramatically (3.83% and 7.81%, respectively).

Keywords: Formaldehyde; Heavy metals; Landfill leachate; UASB

1. Introduction

Landfills are important infrastructures in urban development and serve to effectively store and manage solid waste. The generation of leachate is an unavoidable condition and is a threat to the natural environment [1]. Landfill leachate is made by percolation of precipitation through the landfill waste and by decomposition of the carbonaceous material in the waste. Over time, substantial amounts of leachate are collected at the bottom of the landfill. The leachate may be a probable hazard to the quality of groundwater and may also be toxic to aquatic life [2].

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Leachate is a viscous black or brownish liquid that is rich in organic matter but also typically contains many inorganic chemicals including phosphates, nitrates, and metal salts. Leachate may also contain heavy metals such as As, Fe, Ni, and Cd, which are potentially toxic to the environment, flora and fauna [3]. A typical leachate contains a chemical oxygen demand (COD) of 4,000–20,000 mg L^{-1} depending on the maturity of the leachate [4]. Mature leachate results from previous treatment (biological, chemical, physical or any combination therein) have the characteristic of high COD and a low biochemical oxygen demand (BOD) to COD ratio [5]. The concentration of heavy metals in the mature leachate may be up to four times higher than in fresh leachate due to accumulation [4,5]. Due to the chemical complexity of the leachate, a combination of treatment methods is required. Currently, leachate is treated by biological processes such as aerobic ponds or activated sludge reactors. However, the resulting values of COD and absorbable organic halides are still relatively high [6]. As a result, an alternative treatment method is preferred. Anaerobic treatment technology serves as a suitable alternative and has been used for over a century in the treatment of domestic and industrial wastewaters.

The concentration of heavy metals from various landfills could vary due to, for example, differences in the materials dumped at the landfill site, weathering effects and differences in measurement methods. In Malaysia, the As and Ni concentration exceed the permitted value [6]. Foo et al. [7] and Yusof et al. [8] found out that the amount of Ni and As is higher than the permitted effluent standard. The least promising news is that the leachate sample gets into Malaysian riverine systems [8]. This means that the environment is exposed to heavy metals, which then also could leach into daily diets via the food chain.

Anaerobic treatment methods are more suitable than aerobic treatment systems for concentrated leachate streams due to lower operating costs, emissions of biogas, and low sludge production [9]. Anaerobic treatment is a biological process in which organic matter is degraded into a series of gaseous products – namely, CH_4, CO_2 and H_2. The liquid effluent contains refractive compounds with a significant presence of nitrogen, phosphorus and mineral compounds of K, Ca and Mg. The conversion of organic compounds to methane is a very complex process and requires the presence of different microbial species.

Various anaerobic processes for the treatment of landfill leachate have been studied in recent years such as fluidized bed reactors [10], sequencing batch reactors [11], up-flow anaerobic sludge bed (UASB) reactors [12–16], anaerobic membrane bioreactors [17,18] and other kinds of anaerobic digesters [19]. Among these technologies, UASB has been the most widely investigated in different scales and has been acknowledged as an alternative cost-effective process for the treatment of sanitary wastewater and a wide variety of high-strength industrial wastewaters. UASB is a robust technology and is by far the most widely used high-rate anaerobic process for wastewater treatment [20]. Anaerobic treatment of landfill leachate in UASB reactors may remove 65%–76% of COD and more than 90% of BOD [21].

To date, little work has been done on the treatment of landfill leachate containing heavy metals and formaldehyde (FA). There were a lot of studies done on the treatment of heavy metal by UASB. However, there is still very little work on the treatment of 'matured landfill leachate' by UASB. Most of the previous works were either using synthetic metal wastewater or synthetic mixture heavy metal wastewater, and there were also studies that used industrial metallurgy wastewater. However, once again there was a lack of studies on matured landfill leachate (more toxic compared with fresh leachate as it contains more recalcitrant compounds). Although UASB is widely accepted as an efficient process for landfill leachate treatment, its effectiveness for municipal solid waste (MSW) leachate containing heavy metals and FA need to be reevaluated; this is particularly true since the presence of heavy metals and FA may cause inhibition to anaerobic microorganisms and inhibit the treatment process [22]. This paper explores the feasibility of treating matured leachate from MSW using a UASB reactor that contains heavy metals and FA. Specifically, the effect of increasing organic loading rate (OLR) to the reactor performance was evaluated in terms COD removal, the effect on pH and volatile fatty acid (VFA) and methane concentrations. In addition, heavy metal (As, Fe, Ni and Cd) and FA removal from the leachate was examined.

2. Materials and methods

2.1. Up-flow anaerobic sludge bed

The UASB used in this experimental study was 18 cm in internal diameter (i.d.) and 110 cm in height, with an active volume of 20 L. The reactor had a three-phase separator baffle (pore diameter of 2 mm) placed 2 cm below the effluent ports to prevent floating granules from being washed out with the effluent (Fig. 1). Sampling ports were placed at 8 cm intervals (lowest being 21 cm from the base) that allowed biological solid and liquid samples to be

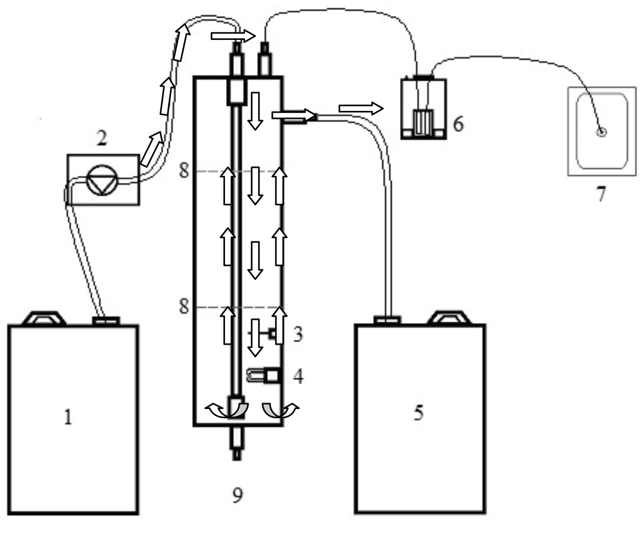

Fig. 1. Schematic diagram of the reactor setup: (1) feed tank, (2) peristaltic pump, (3) temperature sensor, (4) heating element, (5) effluent tank, (6) optical-bubble counter, (7) tedlar gas bag, (8) gas–liquid–solid separator, and (9) UASB.

withdrawn from the sludge bed. The influent wastewater entered through a 2.7 cm i.d. down comer tube in the head plate that extended within 105 cm of the reactor base and allowed feed to flow upward through the sludge bed. A temperature controller and heater were installed to maintain a reactor temperature of 37°C. The UASB reactor can work in a wide range of temperatures, supporting a range of microorganisms from mesophilic to thermophilic species. For this study, a temperature of 37°C was chosen to support mesophilic microorganisms in the treatment of the matured leachate due to its treatment efficiency and lesser energy requirement [23].

2.2. 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 = 2,500 mg L^{-1}, As = 9.40 mg L^{-1}, Fe = 12.8 mg L^{-1}, Ni = 0.50 mg L^{-1}, Cd = 0.43 mg L^{-1} and FA = 8.6 mg L^{-1} (Table 1). The leachate used in this study was collected at once and stored, then used throughout the study. Thus, the characteristics were constant.

2.3. Reactor operations

The reactor was seeded with anaerobically digested sewage sludge (Bunus Sewage Treatment Plant, Kuala Lumpur). On average, 12 L of sieved sludge (using 2.0 mm mesh) was added to the UASB, the remaining volume being filled with tap water. This amount of sludge contributed substantially to the solid requirement in the reactor system after settling. The sieved sludge contains a total solids of 30,100 mg L^{-1} and total volatile solids of 9,525 mg L^{-1}. After seeding, the head plates were attached and the headspace above each compartment was flushed with nitrogen gas to displace residual air in the system before introducing the feed. The reactor was allowed to stabilize at 37°C for 7 d without further modification. The start-up of the reactor was carried out using dilute leachate with a 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 added. The OLR was increased stepwise from 0.125 to 0.625 kg COD m^{-3} d^{-1} at hydraulic retention time (HRT) of 4 d and increased further from 0.833 to 2.5 kg COD m^{-3} d^{-1} by reducing the HRT (Table 2). Finally, the OLR was reduced again to 0.625 kg COD m^{-3} d^{-1} (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 (Bio-Systems Corporation, Asia Pacific Sdn. Bhd, Malaysia) macronutrient supplement. The choice of this nutrient was based on inadequate nutrients in the landfill leachate. There is no excessive nutrients were added to the feed as N100 was first diluted 10 times of its original concentration. In addition, the reactor was operated using these nutrients previously for the treatment of palm oil mill effluent and showed stable reactor operations. Average values of the measured parameters quoted for each OLR were based on three data points taken when the reactor achieved a steady state.

Table 1
Characteristics of leachate

Parameter	Results
pH	8.0
Temperature, °C	26.0
COD, mg L^{-1}	2500
BOD_5 at 20°C, mg L^{-1}	486
Total suspended solids, mg L^{-1}	220
Oil and grease, mg L^{-1}	0.6
VFA, mg L^{-1}	500
Arsenic (As), mg L^{-1}	9.40
Cadmium (Cd), mg L^{-1}	0.43
Formaldehyde (FA), mg L^{-1}	8.60
Iron (Fe), mg L^{-1}	12.80
Nickel (Ni), mg L^{-1}	0.50

Table 2
Summary of reactor operating conditions

Feed COD (mg L^{-1})	OLR (kg COD m^{-3} d^{-1})	HRT (d)	Days
500	0.125	4.0	1–20
1,500	0.375	4.0	20–40
2,500	0.625	4.0	40–60
2,500	0.833	3.0	60–75
2,500	1.250	2.0	75–85
2,500	2.500	1.0	85–90
1,500	0.375	4.0	90–105

2.4. Sampling and VFA analysis

Sample analysis such as COD and pH were conducted according to standard methods [24]. The total biogas volume was determined using an optical gas bubble counter. The biogas composition was determined using a portable gas analyzer (GA2000, Geotechnical Instruments, USA). The FA content was analyzed using high-performance liquid chromatography (1220 Infinity LC, Agilent Technologies, UK) with the following conditions: Zorbax® column (C18, 4.6 mm × 250 mm i.d., 5 μm particle size); mobile phase = 70/30 acetonitrile/water (v/v); flow rate = 1.2 mL min^{-1}; ultraviolet detector operated at 360 nm; and injection volume = 20 μL. Heavy metal analysis of the leachate was conducted using an atomic absorption spectrometry (AA-7700, Shimadzu Corp., Japan).

3. Results and discussion

3.1. pH Levels

Table 3 illustrates the average pH variations in the UASB when the OLR was gradually increased from 0.125 to 2.50 kg COD m^{-3} d^{-1}. The pH levels were generally stable (pH 8.37 to 7.53) in the UASB until the reactor OLR exceeded 0.625 kg COD m^{-3} d^{-1}. For anaerobic digestion, the alkaline phase is preferred to maintain stable microbial populations in the sludge. Many anaerobic reactors

Table 3
Effect of OLR on reactor performance

OLR (kg COD m^{-3} $d^{-1)}$	COD removal (%)	CH_4 composition (%)	VFA (mg L^{-1})	pH	Total volume of CH_4 (L)
0.125	65.70	38.50	157.7	7.69	4.32
0.375	62.30	40.60	86.70	8.37	5.64
0.625	31.17	29.40	64.26	7.53	3.01
0.833	23.70	20.30	120.0	6.77	1.76
1.250	19.70	14.70	156.0	6.10	1.52
2.500	9.33	9.53	255.7	5.48	0.61
0.375	55.57	39.98	157.3	8.37	4.21

fail to operate at low pH (less than 6.7). It is important to maintain a suitable alkalinity in the reactor. The alkalinity was maintained in the reactor at 1,000–2,000 mg L^{-1} as $CaCO_3$ using sodium hydroxide (NaOH). At a reactor OLR of 0.833 kg COD m^{-3} d^{-1}, the pH in the reactor dropped to 6.77 due to the rapid production of VFAs resulting from increased acidogenic activity. A further increase in the OLR to 1.25 and 2.50 kg COD m^{-3} d^{-1} diminished the pH of the reactor to 6.10 and 5.48, respectively. However, when the reactor OLR was reduced back to 0.375 kg COD m^{-3} d^{-1}, the pH in the reactor recovered to 8.37 indicating that the acidogenesis and methanogenesis had recovered to previous levels under low OLR conditions. The pH is an important parameter in anaerobic treatment performance, and many studies have shown the optimum pH for the anaerobic digestion is in the alkaline region [25]; this may be due to the sensitivity of methanogens to acidic conditions. It is also well documented that the cause of the pH drop in anaerobic reactors is due to the VFA production by the acidogenic bacteria. However, Rodriguez et al. [26] demonstrated that acidogenic bacteria produce VFA which cause a pH drop, while methanogenic bacteria removes VFA and causes an pH increase. From the pH data, it can be assumed that the metabolic processes differed between each OLR of the UASB system causing each OLR to favor a unique population of microorganisms.

3.2. Volatile fatty acids

As displayed in Table 3, the VFA concentration in the reactor was lower than 160 mg L^{-1}, when operated at an OLR in the range of 0.125–1.25 kg COD m^{-3} d^{-1}. Increasing the OLR beyond 1.25 kg COD m^{-3} d^{-1} resulted in higher VFA concentrations in the effluent. A drastic increase in VFA concentration was observed (255.7 mg L^{-1}) at OLR of 2.5 kg COD m^{-3} d^{-1}. When the OLR was reduced to 0.375 kg COD m^{-3} d^{-1}, the VFA concentration began to decline and stabilized to 157.3 mg L^{-1}. Reduced contact time between the substrate and biomass in UASB favored the activity of acidogens, leading to decreased methanogen activity in the reactor [27]. VFA concentration is an indicator of feed utilization by anaerobic microorganisms [28]. When there is a build-up of VFA in the anaerobic system, it is likely an indication of the anaerobic microorganisms' failure to utilize the VFA as feed [29]. Some of the variation in the VFA profiles may be influenced by the presence of inhibitory substances such as heavy metals [30].

3.3. COD removal

At an OLR of 0.125 kg COD m^{-3} d^{-1} (HRT 4 d), the average COD removal efficiency was 65.70% (Table 3). This condition was also observed by other researchers during the acclimatization period of anaerobic digestion [31]. Increase of the OLR from 0.375 to 1.250 kg COD m^{-3} d^{-1} resulted in a decreasing COD removal efficiency, until 9.33% was observed at an OLR of 2.50 kg COD m^{-3} d^{-1}. It is unlikely that this was caused by limitations in the UASB reactor as this reactor has been shown to achieve over 90% COD removal at high OLR (e.g., more than 20 kg COD m^{-3} d^{-1}) [32]. However, matured landfill leachate containing a high proportion of recalcitrant and complex organic carbon content may limit 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 [33,34]. Organic matters wash out from the reactor in the form COD may have contributed to the overall low removal of COD.

3.4. Biogas composition

The average biogas concentration fluctuates from 38.50% to 9.53% (Table 3), likely due to the changes in the OLR, since the methanogenic bacteria are sensitive to the changes in feed OLR. As an indirect measure of biomass fluctuations in the reactor, the suspended solids (data not provided) in the reactor correlate well with the methane generation. The biogas concentration profile follows the COD removal efficiency, where the concentration is reduced concomitantly with COD removal. A similar trend was also observed for the VFA profile. VFA decreased when methane composition decreased in an anaerobic treatment process [35]. The methane profile has a close relationship with pH where a decrease in the pH affects the methane generation [36]. Overall, the methane percentage was low probably due to the fact that matured leachate contains less organic fraction. Besides that the low pH could also be a reason behind the lack of methanogenic activity.

3.5. Heavy metal and FA degradation

Fig. 2 shows the heavy metal removal in leachate from the anaerobic digestion via UASB. The figure also shows the comparison of difference in influent and effluent parameters. Generally, it was found that the concentration of heavy metals in the effluent was lower than in the influent. This indicates that the anaerobic microorganisms may have utilized the heavy

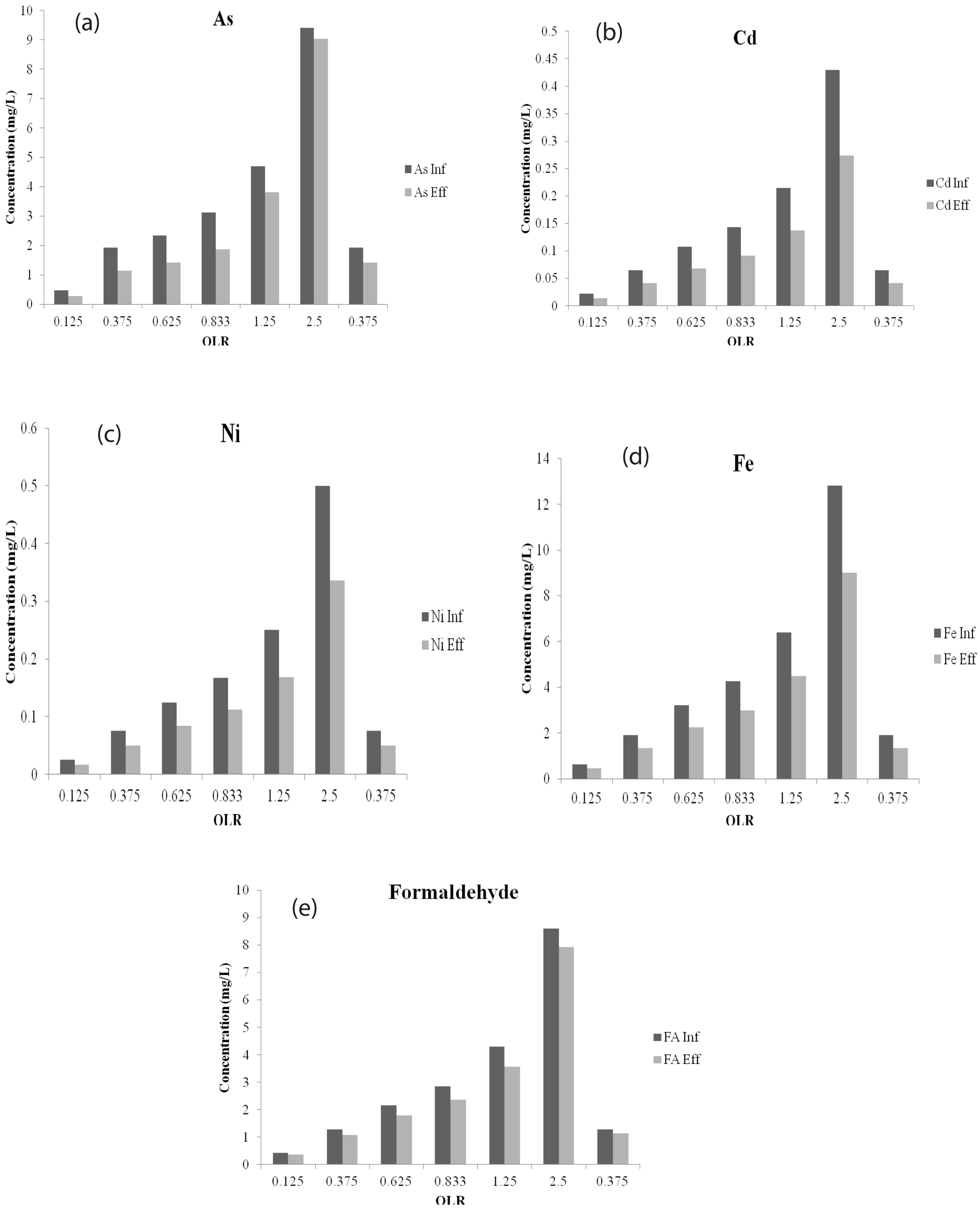

Fig. 2. Concentration of (a) As, (b) Cd, (c) Ni, (d) Fe, (e) FA in influent and effluent of the UASB operated at various OLR.

metals or the metals were accumulated in the sludge. Dong et al. [37] observed that during anaerobic digestion, the concentration of heavy metal in the sludge increased. Thanh et al. [38] found that methanogenic bacteria require trace elements such as Fe and Ni for anaerobic digestion and methanogenic activity, supporting the idea that the metal concentrations may drop due to utilization by the microbiota. Cestonaro do Amaral et al. [39] demonstrated that the metal retention capacity of bioreactors decreased while other metal fractions remained the same or decreased when OLR was increased. The above studies demonstrate that the heavy metal removal during the anaerobic digestion may vary and may depend on the reactor configuration, sludge washout type of wastewater and operating conditions. In the current study, the heavy metal removal profile fluctuates according to the type of the heavy metal. Table 4 shows the effect of OLR on the heavy metal removal. It can be seen that the removal of Cd, Ni and Fe was almost constant regardless of the OLR (around 36% for Cd, 32% for Ni and 29% for Fe). This shows that when the OLR was increased from 0.125 to 2.50 kg COD m^{-3} d^{-1} and then decreased to 0.375 kg COD m^{-3} d^{-1}, the heavy metal removal efficiency for Cd, Ni, and Fe were almost consistent. The stable population of bacteria appears to tolerate the introduction of these metals into the reactor system when the OLR gradually increased from 0.125 to 2.5 kg COD m^{-3} d^{-1} and decreased back to 0.375 kg COD m^{-3} d^{-1}. Contrarily, As and FA displayed a degree of removal at low OLRs (e.g., 40% and 17% at 0.125 and 0.833 kg COD m^{-3} d^{-1}, respectively), but at high OLR (e.g., 2.5 kg COD m^{-3} d^{-1}), both As and FA decreased dramatically (3.83% and 7.81%, respectively). When the OLR was decreased back to 0.375 kg COD m^{-3} d^{-1}, some removal was still evident (around 26% and 12%, respectively), signifying that the microorganisms in the reactor were capable of recuperating from the shock load. Zhao et al. [40] reported that FA can also aid in the removal of As; however, the significant contribution of FA towards As removal in the current study was not investigated.

Microorganisms are known to have the ability to bind with metals including toxic heavy metals [41]. A study by Lu and Hegemann [42] demonstrated the inhibition of anaerobic bacteria at FA concentrations of 200 mg L^{-1}. In a separate study carried out by Vidal et al. [43], the toxic level of FA was determined to be 100 mg L^{-1}. It is possible that even with a relatively low leachate concentration of FA (8.6 mg L^{-1}), accumulation may occur causing FA concentrations to be high in the sludge at OLR of 2.50 kg COD m^{-3} d^{-1} and may have contributed to the poor performance of the UASB reactor. Although actual FA concentrations in the UASB were not measured, this type of accumulation has been noted previously [40]. From the results (Fig. 2), it is visible that there was some form of removal of the heavy metals and could have occurred by either bioaccumulation or accumulation in sludge [44]. Bioaccumulation was less likely to be possible thus the pollutants might have accumulated in the sludge.

According to Xie et al. [45], acidification (acidogenesis and acetogenesis) was dominant in an anaerobic bioreactor treating leachate due to pH decrease (7.83–6.15) resulting from VFA production. They pointed out that the lower pH resulting from acidification enhances metal solubility and leaching for Zn, Cu, Cd, Pb and Ni. The solubility of heavy metals in landfill leachate is affected by the concentrations of soluble COD (sCOD) [45]. Low-molecular-weight organic compounds that make up the sCOD can form soluble complexes with heavy metals, which prevent metals adsorbing or complexing to the solid phase in MSW, thereby increasing dissolved metal concentrations as metal-dissolved organic matter complexes [46]. In the current study, the sCOD of the leachate varied greatly (500–2,500 mg L^{-1}). It is highly probable that components of this dissolved organic matter contributed to greater overall solubility of heavy metals in the leachate. However, further research on the simulation of metal and dissolved organic matter complexation during the entire course of anaerobic digestion of landfill leachate should be performed [47].

Table 4
Effect of OLR on heavy metal and FA removal

OLR (kg COD m^{-3} d^{-1})	Removal (%)				
	As	Cd	Ni	Fe	FA
0.125	40.00	36.36	32.00	29.69	17.21
0.375	40.00	36.92	33.33	29.69	17.13
0.625	40.00	37.04	32.80	29.69	17.12
0.833	40.00	36.36	32.93	29.69	17.13
1.250	18.72	36.28	32.80	29.70	17.12
2.500	3.83	36.28	32.80	29.70	7.81
0.375	26.04	36.92	33.33	29.69	12.25

4. Conclusions

This study demonstrates that the UASB reactor can be used to treat matured landfill leachate; however, the treatment efficiency of the reactor was negatively affected at an OLR of 2.5 kg COD m^{-3} d^{-1}, probably due to the inhibition by heavy metal and FA in the leachate at high OLR. At high OLR, the concentration of heavy metals and FA may have increased due to accumulation, leading to methanogen inhibition. The removal of Cd, Ni and Fe was nearly constant regardless of the OLR; however, Fe and FA removal was affected at higher OLR. FA has been reported to be a strong inhibitor of all microorganisms involved in anaerobic degradation, although no information about its inhibition pattern has been given so far, especially in the treatment of landfill leachate. It is recommended that the microbial activity of the reactor sludge to be evaluated in future study to determine the correlation of heavy metal and FA removal by different microorganisms.

Acknowledgments

The authors desired to show the highest gratitude to Mr. Azmi Abu Bakar who helped with the apparatus and materials for the experiments and also, special acknowledgment to Universiti Teknologi Malaysia for funding this study. The authors also thank the Ministry of Education Malaysia for the Research University Grant (Vote number: Q.K130000.2510.13H11).

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