Iran. J. Environ. Health. Sci. Eng., 2011, Vol. 8, No. 1, pp. 3-14

AEROBIC THERMOPHILIC TREATMENT OF LANDFILL LEACHATE IN A MOVING-BED BIOFILM BIOREACTOR

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Received 20 April 2010; revised 27 October 2010; accepted 18 December 2010

ABSTRACT
In the present study the landfill leachate obtained from Tabriz Landfill site was first treated in a suspended growth activated sludge bioreactor which then was turned to a moving-bed biofilm bioreactor under thermophilic conditions (50ºC). The average Soluble Chemical Oxygen Demand (SCOD) removal ranged from 70% to 62% as Organic Loading Rate (OLR) increased from 0.454 to 4.678 kgCOD/(m^3.d). Hydraulic Retention Time (HRT) values were set to 12, 14, 16.8, and 21 h. The recycle ratio of biomass ranged from 1 to 1.5. The total biomass concentration was in the range of 2600-3100 mg/L of which 60-65 percent was in the form of biofilm. It was COD removal efficiency was 5% to 10% higher in moving-bed biofilm operation compared to suspended growth one at the same OLR and HRT. Respirometry experiments revealed that readily biodegradable organic compounds accounted for 5.7% and 5.2% of substrate under thermophilic and mesophilic conditions, respectively. The maximum resulting oxygen uptake rates were 0.21 mgO_2/(L.min) and 0.16 mgO_2/(L.min) under thermophilic and mesophilic conditions, respectively.

Key words: Landfill leachate; Thermophilic; Aerobic; Moving bed biofilm bioreactor

INTRODUCTION
Municipal Landfill leachates contain high concentrations of organic content (values of COD vary from 100 to 70900 mg/L), high amounts of ammonia (ranging from 0.2 to 13000 mg/L) and refractory organic materials (Renoua *et al.*, 2008). The ratio of BOD_5/COD, decreases rapidly with the aging of the landfills (Alvarez-Vazquez *et al.*, 2004). The composition of leachates obtained from different sanitary landfills shows a wide variation. Factors determining the quality of such leachates include the age of landfill, precipitation, seasonal weather variation, waste type and composition (Wintgens *et al.*, 2003). Particularly, the age of landfill has a significant effect on the composition of landfill leachate (Baig *et al.*, 1999).

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Considering the high organic content of landfill leachates and the limitations of suspended growth activated sludge systems such as inadequate sludge settleability (Loukidou and Zouboulis, 2001) and excess sludge production (Hoilijoki *et al.*, 2000), it seems promising to use more advanced processes like moving-bed biofilm bioreactors which benefit from both attached-growth and well mixing of the liquor (Loukidou and Zouboulis, 2001). Moving-Bed Biofilm Bioreactors (MBBRs) have been studied for the treatment of thermomechanical pulping whitewater (Jahren *et al.*, 2002), pulp and paper mill effluent (Suvilampi and Rintala, 2004), phenolic synthetic wastewater (Hosseini and Borghei, 2005), seawater (Dupla *et al.*, 2006), landfill leachate (Chen *et al.*, 2008; Kulikowska *et al.*, 2009), synthetic wastewater (Yang *et al.*, 2009) and synthetic wastewater aiming at biological nitrogen compounds removal through partial nitrification-denitrification process (Zafarzadeh *et al.*, 2010) . MBBR process is based on the use of suspended porous polymeric carriers, kept in continuous movement in the aeration tank, while the active biomass grows as a biofilm on their surfaces. Researchers have proven that MBBR systems possess advantages such as high biomass concentration in the bioreactor, high organic loading rates, great tolerance to loading shocks, relatively less reactor size and no sludge bulking problem (Chen *et al*., 2008).

Aerobic thermophilic treatment offers several advantages such as rapid biodegradation rates, low sludge yields, rapid inactivation of pathogenic microorganisms, high loading rate thus reducing the retention time for treatment and capital cost, and tolerating varying operational conditions. However, thermophilic processes have also their disadvantages such as poor flocculation and settleability of biomass, high turbidity of effluent, foam formation and need to an external energy source (Lapara and Alleman, 1999; Suvilampi *et al*., 2005).

The objective of the present study was to evaluate the treatability of landfill leachate under thermophilic conditions by using an aerobic moving-bed biofilm bioreactor. High loading shocks were tested for evaluation of the effectiveness and stability of the whole system. The fractions of readily and non-biodegradable content of landfill leachate were also determined.

MATERIALS AND METHODS

Characteristics of landfill leachate and aerobic activated sludge

Landfill leachate used for the treatment was obtained from the landfill site of Tabriz, Iran. The landfill leachate samples collected at two times had different characteristics as shown in Table 1. Aerobic activated sludge used as inoculum was obtained from the recycle flow exiting the clarifier of Tabriz Wastewater Treatment Plant (WWTP). The characteristics of this activated sludge are summarized in Table 2.

Experimental set-up

Suspended-growth activated sludge bioreactor

The continuous system, as shown in Fig. 1, consisted of an aeration tank with a rectangular cross section which has a length, width, and height of 60, 40, and 40 cm, respectively. Aeration tank was followed by a clarifier with a trapezoid cross section. The lengths of parallel sides are 10 and 40 cm and the height of clarifier is 40 cm. The total and working volumes of aeration tank was 96 and 84 L, respectively. The active volume of clarifier was 35 L. A baffle was located inside the reactor at 50 cm from the front side and 10 cm from the bottom of the aeration tank. The reactor was made of galvanized iron with a thickness of 6 mm. Two air diffusers were installed at the bottom of the reactor to aerate and suspend the biomass. The air flow provided by a central compressor system had a constant rate of 0.4 vvm so that the concentration of soluble oxygen remained at 3 to 4 mg/L. Heating of the liquor was done by hot

Table 1: Characteristics of landfill leachate obtained from Tabriz landfill site

No.	pH	BOD_5/COD	COD	TS	TDS	TVS	TSS	VSS
1	6.7±0.06	0.59±0.04	35600±1060	41100±1520	32000±660	14800±330	9100±140	2800±90
2	7.2±0.06	0.57±0.02	12200±440	12600±360	4360±560	10000±820	2600±260	650±130

Notes: All units are in mg/L except for BOD5/COD and pH

The values represent the average value ± standard deviation

Table 2: Characteristics of activated sludge obtained from the recycle flow of Tabriz WWTP

	TSS	VSS	FSS
Activated sludege	8300±230	5700±110	2600±80
Concentrated Activated Sludge	15700±70	11350±100	4350±110

Note: All units are in mg/L

The values represent the average value ± standard deviation water tubes enclosing the reactor. Temperature was controlled by an on/off controller. The feed (leachate) was fed to the bioreactor by means of a reciprocating pump from a 60 L tank. The recycle flow was returned to aeration tank by another reciprocating pump. The fittings were made of silicon. The bioreactor was covered to minimize the effects of evaporation and stripping.

Moving-bed biofilm bioreactor

The physical appearance of the bioreactor in attached-growth operation was almost the same as suspended-growth one. About 54% of the reactor volume was filled by plastic carriers. About 14 L of reactor volume was reduced by blocking a portion of bioreactor inside (from the baffle zone) to prevent the carriers from leaving the aeration tank. So, the total working volume of bioreactor including the biofilm amounted to approximately 70 L.

In order to use an available and cheap carrier with outside surface area and a density close to water, rubber hose tubes cut into ring like parts were used. The diameter and length of the carriers were 17 and 14 mm, respectively. The estimations showed that around 7865 particles with an approximate 250 m^2/m^3 surface area filled the bioreactor.

Operating procedure

Non-biodegradable fraction of landfill leachate

To find the non-biodegradable fraction of feed (landfill leachate) 0.2 L of mixed liquor containing thermophilic and mesophilic biomass were poured in separate beakers and 0.4 L of feed was added to each of them. Each beaker was aerated for 3 days under mesophilic and thermophilic conditions. The ratio of remaining COD to feed COD shows the non-biodegradable fraction.

Respirometry experiments

The purpose of respirometry experiments is to evaluate readily biodegradable content of feed (Spanjers *et al*., 1995). A double-walled glass container with an effective volume of 125 mL was used and isolated from the environment by an oxygen meter probe installed at the top. Some mesophilic and thermophilic biomass were taken and aerated in separate containers to get them activated and reached the endogenous respiration, meaning that their oxygen uptake reached a constant value. Then, a medium containing minerals of NH_4Cl (480 mg/L), K_2HPO_4 (140 mg/L), $MgSO_4.7H_2O$ (50 mg/L), $FeSO_4.7H_2O$ (2.5 mg/L), and $CaCl_2.2H_2O$ (2.5 mg/L) was added to the container. Next, 5 mL of wastewater with COD of 1200 mg/L was added to the container and the values of Dissolved Oxygen (DO) was recorded every one minute. The oxygen uptake rate (OUR) curve versus time was then prepared. The area below the curve, after subtracting the area related to endogenous respiration from it, shows Short-Term Biochemical Oxygen Demand (BOD_{st}).

The readily biodegradable content by substituting the values of BOD_{st} and biomass Yield (Y) into equation 1 and solving it (Petersen *et al*., 2002).

$$Y = 1 - \frac{BOD_{st}}{COD_{readily}} \qquad (1)$$

Batch experiments

Batch experiments were conducted to find the effect of evaporation and stripping on the thermophilic treatment and COD reduction. T1 L beakers filled by 500 mL of feed were used and covered by aluminum foils. For aeration, stone diffusers were used. The beakers were put in a waterbath of constant 50ºC temperature while being aerated for 20 h.

The non-biodegradable fraction of wastewater was also determined by batch experiments, which were composed of 0.2 L mixed liquor and 0.4 L feed. They were aerated for about 3 days at two mesophilic and thermophilic conditions.

Continuous suspended-growth operation

To activate thermophilic biomass, after 9 days operation of the system under mesophilic conditions (40°C), the temperature of the system was increased from 40°C to 50°C within 7 days. The changes in Mixed Liquor Suspended Solids (MLSS) and Mixed Liquor Volatile Suspended Solids (MLVSS) with time are presented in Fig. 2. Since the concentration of activated thermophilic biomass was low, some fresh biomass was added to the system and let it operate for 24 h in batch mode at 50°C. The changes in MLSS and MLVSS with time are presented in Fig. 3. After reaching an acceptable concentration of biomass in the bioreactor, the feed was introduced to the bioreactor at HRT of 12 h. The OLR increased gradually. After day thirteen the suspended-growth operation changed to biofilm operation because of the increase in biomass in the effluent. The main parameter for observing the operation of reactor was the changes of influent and effluent COD. The changes in MLSS and MLVSS were also recorded.

Continuous attached-growth operation

The operation of bioreactor was observed at different HRTs (12, 14, 16.8, and 21 h). The influent loading rate was increased gradually to find the maximum loading capacity and optimum HRT and influent organic loading rate of the system. The whole period of operation was around 45 days.

Analytical methods

The values (TSs), (TSS), T(TDS), (TVS), (FSS), (VSS), (VDS), COD, and 5-day Chemical Oxygen Demand (BOD_5) of samples were measured according to the sections 2540 B, 2540 D, 2540 C, 2540 E, 2540 G, 2540 G, 2540 G, 5220 C, 5210 B of the methods given in Standard Methods (APHA, 1998), respectively. The pH values of the samples were measured by a HANNA® pH meter electrode.

RESULTS

Evaluation of non-biodegradable fraction of landfill leachate

The sample leachate COD before and after treatment was 4350 mg/L and 1527 and 1117 mg/L under thermophilic and mesophilic conditions, respectively. It means that the non-biodegradable fraction is 0.35 and 0.26 for these two conditions.

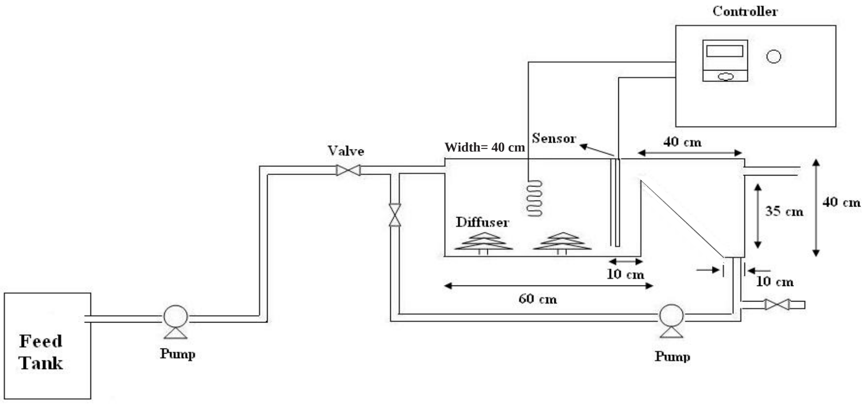

Fig.1: Schematic diagram of the continuous bioreactor

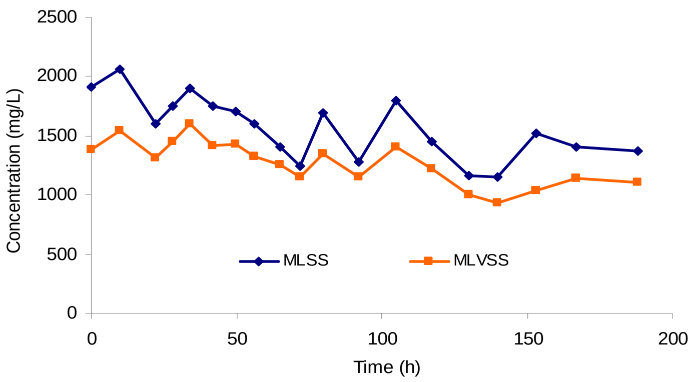

Fig.2: Changes in biomass concentration during the temperature rise

Evaluation of oxygen uptake rate

As illustrated in Figs. 4 and 5, the maximum oxygen uptake rates were 0.21 and 0.16 mgO_2/L.min under thermophilic and mesophilic conditions, respectively. The resulting values for easily-biodegradable, slowly-biodegradable, and non-biodegradable (inert COD) content were 5.2%, 68.8%, and 26.0% at 30^0C and 5.4%, 59.6%, and 35.0% at 50^0C, respectively.

Respirometry results

Figs. 4 and 5 show the OUR values with respect to time. Fig. 6. represents the DO reduction with time under thermophilic conditions. The dashed lines in these figures show the average endogenous respiration which was estimated as 0.05 and 0.02 mgO_2 /L min for thermophilic and mesophilic conditions, respectively. BOD_{st} was calculated to be 48 mg/L and 39 mg/L for thermophilic and mesophilic conditions, respectively.

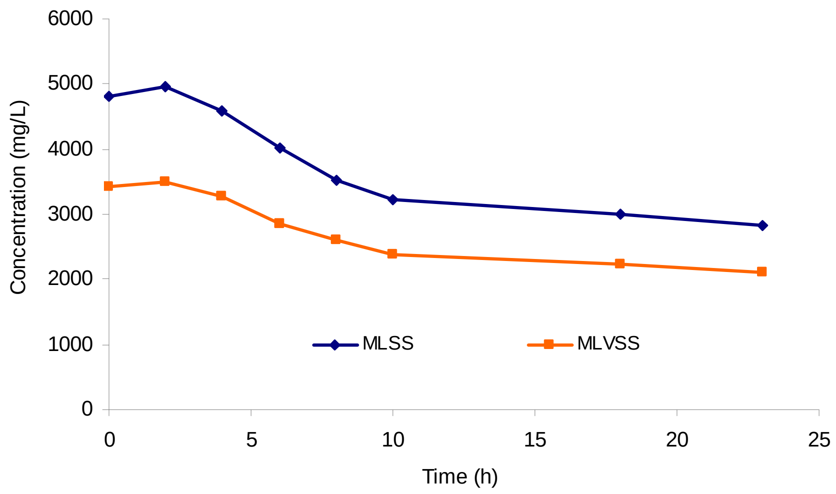

Fig. 3: Changes in biomass concentration during the aeration of activated sludge at 50ºC

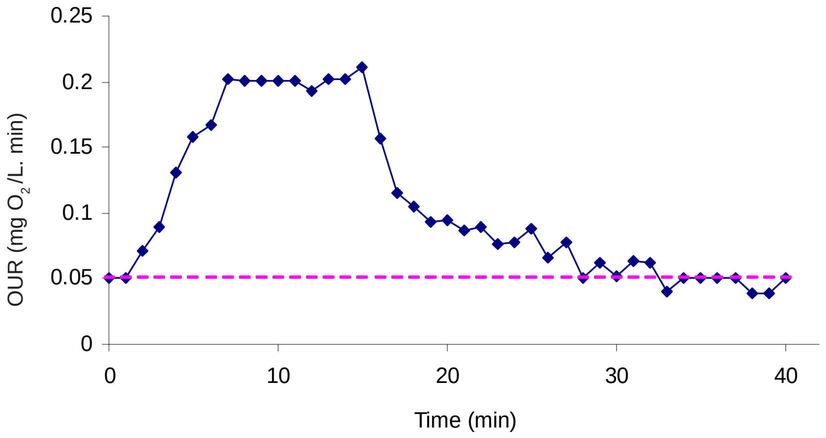

Fig. 4: OUR changes with time under thermophilic conditions

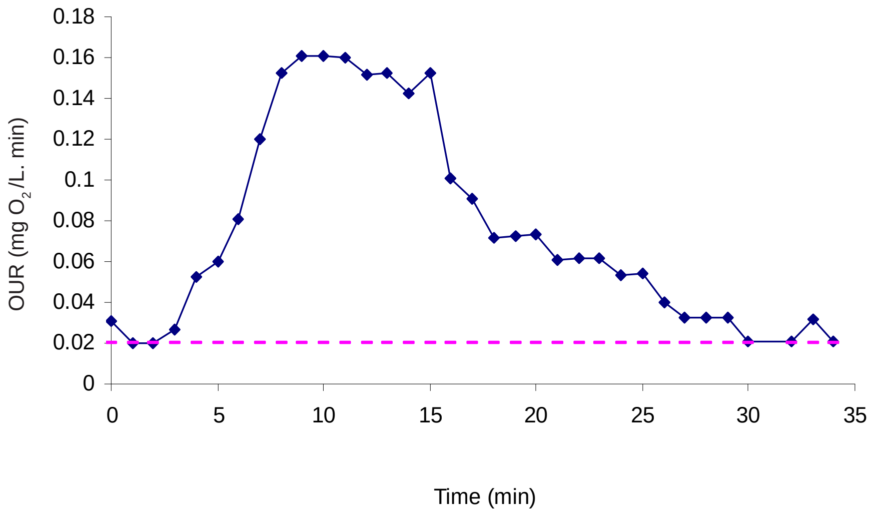

Fig. 5: OUR changes with time under mesophilic conditions

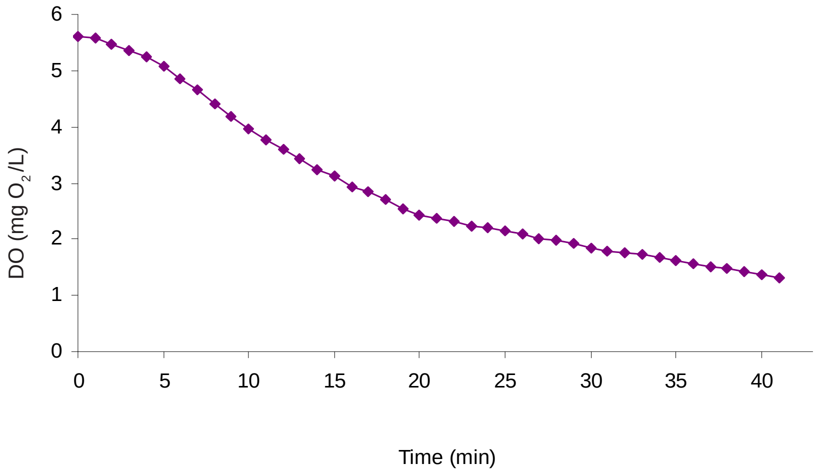

Fig. 6: DO reduction with time under thermophilic conditions

We assumed an average value of 0.35 mgSS/mgCOD for Y at mesophilic conditions and 0.30 mgSS/mgCOD at thermophilic conditions (Vogelaar *et al*., 2003). The resulting values for readily biodegradable content ($COD_{readily}$) are 69 and 62 mg/L accounting for 5.7% and 5.2% of substrate at thermophilic and mesophilic conditions, respectively.

Batch experiments results

The initial COD of feed which was added to 1 L beakers for determining the effect of temperature on COD and volume reduction was 10500 mg/L. The final COD after 20 h aeration was 10000 mg/L which showed 5% decrease in COD. The volume decrease was 5.2% in this period.

In the second part of batch experiments the initial COD of mixture was 4350 mg/L. The final COD of mixture after 3 days aeration were 1527 and 1117 mg/L under thermophilic and mesophilic conditions, respectively. As a result, the non-biodegradable and inert fraction of feed were estimated to be 35% and 26% at these conditions.

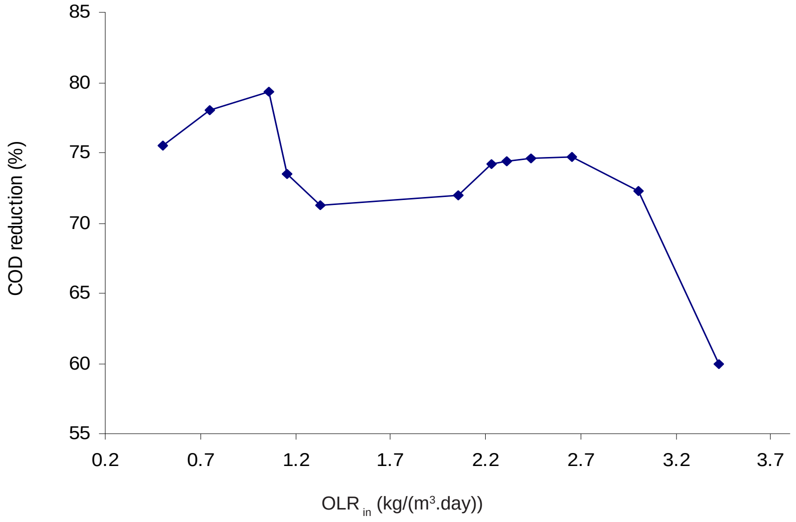

Fig.7: COD removal efficiency versus OLR in activated sludge bioreactor

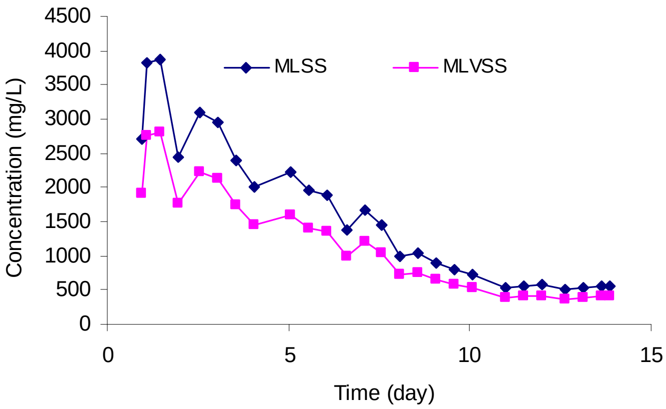

Fig. 8: Solids concentration versus time in activated sludge bioreactor

Performance of suspended-growth activated sludge bioreactor

The initial OLR was 0.5 kgCOD/(m^3.d) which increased to 3.4 kgCOD/(m^3.d) during 14 days of operation. The SCOD removal rate varied from 60% to 79% with an average value of 73%. The organic loading removal rate was in the range of 0.12 kgCOD/(m^3.d) to 1.37 kgCOD/(m^3.d).

The trend of MLSS changes, as illustrated in Fig. 8, shows that MLSS decreases with time to around 500 mg/L mainly due to the increase in OLR.

Performance of moving-bed biofilm bioreactor

The changes in suspended and attached SS and VSS are presented in Figs. 9 and 10. The total solid concentration of biomass (suspended and attached) was in the range of 2600-3100 mg/L, of which 60-65 percent was in the form of biofilm. The average ratios of VSS/SS in the effluent and biofilm were 0.66 and 0.45, respectively. As illustrated in Fig. 11, the COD removal efficiency increases primarily with increase in OLR but it decreases gradually and reaches to a constant value at OLR of about 6 kgCOD/(m^3.d). More increase in OLR (to about 11 kg COD/(m^3.d)) results in decrease in efficiency. The average SCOD removal ranged from 70% to 62% as OLR increased from 0.454 to 4.678 kg COD/(m^3.d).

The typical results of the system are summarized in table 3. The pH of influent and effluent was in the range of 6.5 to 7.5 and 8.7 to 8.9 respectively. The pH of mixed liquor was about 8.3.

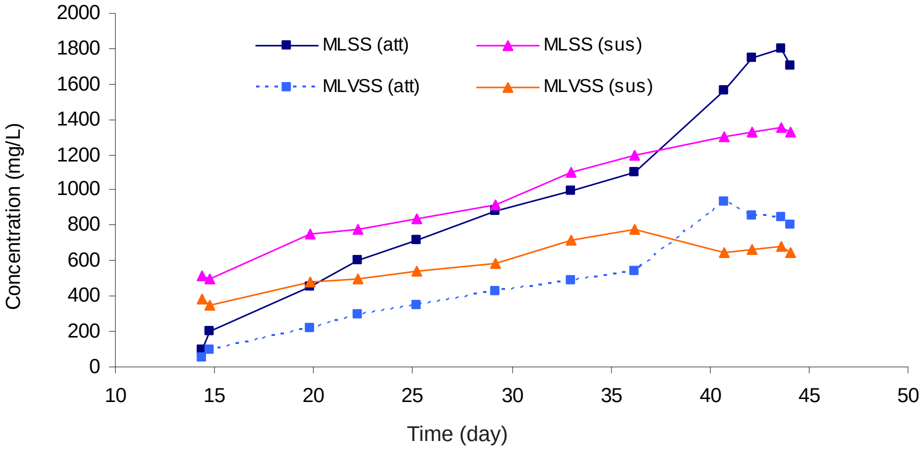

Fig. 9: Suspended and attached solids concentration versus time in MBBR

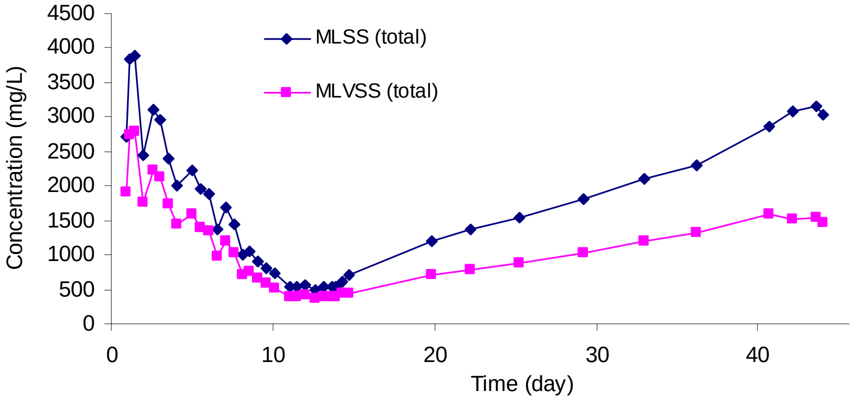

Fig.10: Total solids concentration versus time in MBBR

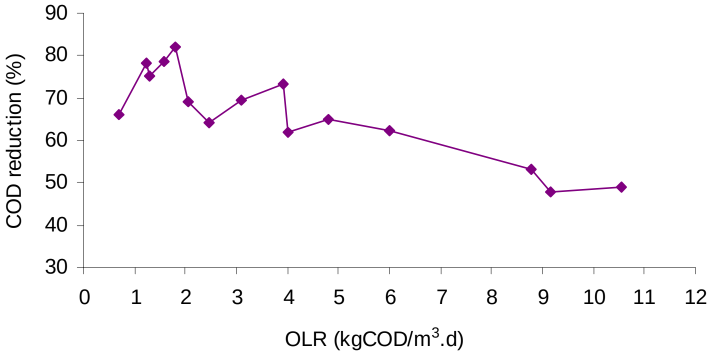

Fig.11: COD removal efficiency versus OLR in MBBR

Table 3: The typical results of thermophilic moving-bed biofilm bioreactor

Time period (day)	HRT (h)	Influent SCOD (mg/L)	Effluent SCOD (mg/L)	OLR (kgCOD/(m^3.d))	Removal rate (kgCOD/(m^3.d))	SCOD removal efficiency (%)	Recycle ratio
1.6	12	1350±80	357±45	1.16±0.09	0.85±0.03	73.5±3.9	1.33
6.6	12	2700±370	690±138	2.31±0.11	1.72±0.06	74.4±8.1	1.33
11.5	12	3510±230	905±150	3.00±0.08	2.23±0.07	74.2±2.6	1.33
26.4	14.8	2890±50	624±94	1.24±0.10	0.86±0.05	78.4±3.1	3.27
27.1	14	2890±50	1030±176	2.48±0.09	1.53±0.07	64.3±5.4	1
36.5	16.8	12300±120	5750±255	8.78±0.07	4.68±0.12	53.3±1.7	1
41.0	16.8	11900±270	8000±84	6.80±0.08	2.23±0.04	32.8±1.1	1.5

Note: The values represent the average value ± standard deviation.

DISCUSSION

In the present study we treated landfill leachate in a suspended-growth activated sludge which then turned to a moving-bed biofilm bioreactor under thermophilic conditions.

Meanwhile, we did some experiments to identify the non-biodegradable and bio-degradable fractions of the landfill leachate.

The maximum respiration rate, which would be achieved during the degradation of readily biodegradable compounds (here as readily biodegradable COD ($COD_{readily}$)) was almost the same for both mesophilic and thermophilic conditions. The reason is that contrary to a higher growth rate for thermophilic biomass, the difference between the ratios of active biomass over the total biomass causes a similarity between maximum respiration rates (Vogelaar *et al.*, 2002).

The difference between non-biodegradable COD at mesophilic and thermophilic conditions could be as a result of (Vogelaar *et al.*, 2002):

1. Thermophilic bacteria are not able to degrade a large group of compounds which can be degraded by mesophilic biomass
2. Thermophilic biomass produces more soluble microbial products SMP in comparison to mesophilic one. Soluble microbial products (SMP) are defined as soluble cellular components that are released during biomass decay (utilization associated products (UAP)) or substrate metabolism (biomass associated products (BAP)) (Laspidou and Rittmann, 2002). Lapara and Alleman (1999) stated that the soluble microbial products at thermophilic conditions contain more fraction of non-biodegradable compounds compared to mesophilic conditions. As mentioned earlier, the percent of readily biodegradable substances which contains simple substrates is the same under mesophilic and thermophilic conditions but when the substrate structure becomes more complicated its degradability declines at thermophilic conditions compared to mesophilic conditions.

Low values of BOD_{st} show that the majority of readily biodegradable content of landfill leachate which mainly consists of volatile fatty acids had been already removed through landfilling process.

While the biomass concentration inside the bioreactor decreased, its content increased in effluent of the bioreactor. The main reason could be the poor sedimentation of thermophilic biomass which can not be coagulated well so that remains mostly dispersed in the liquor and exits the bioreactor (Lapara *et al.*, 1999). To prevent the system from a dramatic efficiency fall, we changed the configuration to moving-bed in order to benefit from suspended and attached growth at the same time.

Microorganisms grow on the surface of carriers and their concentration increases until it reaches a constant value which is around 1700-1800 mg SS/L. High percentage of attached biomass (biofilm) in the MBBR implies that the low concentration of biomass inside the bioreactor is not related to poor immobilization of bacteria on the carriers but it is more likely caused by the limitation of nutrients like nitrogen and phosphorus and the presence of inhibitors in landfill leachate (Park S. *et al*., 2001).

As it can be seen in Fig.7, the process shows good performance at low OLR, but it declines as the OLR increases. Maximum loading capacity of a bioreactor is defined as the loading rate at which the removal rate does not increase with OLR. It can be inferred from Fig.11, ignoring the primary fluctuations, that the maximum loading capacity could be around 6 kg COD/(m^3.d), because the removal rate decreased after this point. But we can see that by increasing OLR to around 10.54 kg COD/(m^3.d) the removal still increased. So the maximum loading capacity would be higher than 10.54 kg COD/(m^3.d).

The results show that increase of HRT does not have a significant effect on COD removal efficiency. HRT within the range of 12 to 16.8 h was appropriate for the system. However, HRTs less than 12 h could decrease the system efficiency. For example, the removal efficiency was 62% at HRT of 9.5 h and OLR of 4 kg COD/(m3.d), while it increased to 73.2% when HRT increased to 14 h. HRT values greater than 15 h did not have a great effect on the efficiency.

In a similar research, Chen *et al*. (2008) used a moving-bed biofilm reactor (MBBR) system with an anaerobic–aerobic arrangement. The contribution of the anaerobic MBBR to total COD removal efficiency reached 91% at an organic loading rate (OLR) of 4.08 kg COD/(m^3.d) at HRT of 4 days, and gradually decreased to 86% when feed OLR was increased to 15.70 kg COD/(m^3.d) at HRT of 0.5 days. The total COD removal efficiency of the system had a slight decrease from 94% to 92% even though the feed OLR was increased from 4.08 to 15.70 kg COD/(m^3.d). A list of previous studies (comprising studies conducted by Welander and Henrysson, 1998; Ødegaard and Leiknes, 2001; Loukidou and Zouboulis, 2001; Jahren *et al*., 2002; Suvilampi and Rintala, 2004; Hosseini and Borghei, 2005 and Yang *et al*., 2009) is shown in Table 4.

Table 4: Different moving-bed biofilm bioreactors used for wastewater treatment

Feeding				Operational conditions				Removal efficiency	
COD (mg/L)	BOD_5/COD	pH	From	Reactor Volume (L)	HRT (h)	OLR (kg COD/(m^3.d))	T (ºC)	(%)	References
800-12300	0.57-0.59	6.5-7.5	Landfill	70	12-21	0.45-4.68	50	62-70 SCOD	Present study
800–2000	-	-	Landfill	5000	17	4	20	20 COD	(Welander and Henrysson, 1998)
219±66	-	7.5±0.1	Municipal wastewater	200	0.33-0.5	30-45	10-15	85-90 COD	(Leiknes and Ødegaard, 2001)
5000	0.2	>7.5	Landfill	8	20-24	5-6	-	81 COD	(Loukidou and Zouboulis, 2001)
2100-2400 (SCOD)	0.5	4.5-4.9	Thermomechanical pulping whitewater	8.55	13-22	1.8-3.8 (kg SCOD/(m^3.d))	55	60-65 COD	(Jahren *et al*., 2002)
-	-	-	Settled pulp and paper mill effluent	-	13±5	2.7±0.9 (kg COD_{filt} /(m^3.d))	46-60	85 COD_{filt}	(Suvilampi and Rintala, 2004)
800	-	7-7.2	Synthetic wastewater	22	8-24	0.8-2.4	23	70-96 COD	(Hosseini and Borghei, 2005)
385-874	-	7.6-8.5	Synthetic wastewater	30	12	0.77-1.75	25	95.6 COD	(Yang *et al*., 2009)

A comparison between biofilm and suspended growth processes at same concentrations and HRTs shows a higher efficiency for biofilm one. For example, the efficiencies of suspended growth and moving-bed bioreactor were 69.3% and 75.3% for influent COD of 1500 mg/L and HRT of 12 h, respectively. It was seen that COD removal efficiency was generally 5% to 10% higher in the moving-bed biofilm operation compared to suspended growth one at the same OLR and HRT.

Since the pH of the liquor was about 8.3, there was no need to any pH control strategy. Even if landfill leachate was acidic, the pH of liquor would approach to neutral because the ammonium concentration is likely to increase in the absence of nitrification (Lapara *et al.*, 1999).

ACKNOWLEDGEMENTS

The authors would like to express their great appreciations to Sahand University of Technology and Tarbiat Modares University for supporting the present research work.

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