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Desalination 204 (2007) 8–16

DESALINATION

www.elsevier.com/locate/desal

Landfill leachate treatment using thermophilic membrane bioreactor

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Received 11 January 2006; accepted 25 February 2006

Abstract

This study was undertaken to investigate the performance of aerobic thermophilic membrane bioreactor (MBR) treating raw landfill leachate from two landfill sites in Thailand (Pathumthani site and Ram Indra site). The leachates from these sites were mixed in different proportions to produce a BOD/COD ratio of 0.39, 0.57, and 0.65, which was investigated in 3 experimental runs. The COD, ammonia, and TKN composition of the mixed leachate was 12,000, 1700 and 1900 mg/L, respectively. BOD was supplemented with glucose and soy protein. The system was operated at 45°C and at a hydraulic retention time (HRT) of 24 hrs. The membrane used was a ceramic membrane with an "outside-in" flow mode and consisted of 22 open fibres with an inner diameter of approximately 2 mm. The COD removal rate increased from an average value of 62–79% while ammonia removal efficiency decreased from 75 to 60% with gradual increase in BOD. Furthermore, a high BOD removal efficiency (97–99%) was also observed. This clearly indicates that thermophilic system is highly suitable for COD and BOD removal especially at elevated organic loading. However, the system does not favor high nitrogen content wastewaters as the ammonia removal efficiency dropped with increasing BOD/COD ratio. Similar trends were found in TKN analysis as well. However, this system could serve as a pretreatment in removing ammonia. The concentrations of soluble and bound extra-cellular polymeric substances (EPS) found in thermophilic MBR were higher when compared to the corresponding concentrations in a mesophilic MBR, which led to a higher rate of fouling in the thermophilic membrane.

Keywords: Extra-cellular polymeric substances; Landfill leachate; Membrane bioreactor; Mesophilic; Thermophilic

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Presented at EuroMed 2006 conference on Desalination Strategies in South Mediterranean Countries: Cooperation between Mediterranean Countries of Europe and the Southern Rim of the Mediterranean. Sponsored by the European Desalination Society and the University of Montpellier II, Montpellier, France, 21-25 May 2006.

1. Introduction

Municipal solid waste disposal in the landfill is the most common solid waste management practice followed throughout the world. However, landfill requires a close environmental engineering surveillance in its design and operation, as it is likely to generate leachate, which would potentially contaminate nearby groundwater and surface water. With the changing nature of domestic refuse composition over the years, the proportion of refuses available for decomposition has greatly increased and thus the organic strength of the leachate, resulting in its greater potential for pollution.

Landfill leachate can broadly be defined as the liquid produced from the decomposition of waste and infiltration of rainwater in the landfill. Generation of leachate occurs when moisture enters the refuse in a landfill, dissolves the contaminants into liquid phase and becomes sufficient to initiate a liquid flow. Leachate varies from one landfill to another with fluctuations that depend on short and long-terms due to variations in climate, hydrogeology and waste composition [1]. Due to this, improvements in landfill engineering are aimed at reducing leachate production, collection and treatment prior to discharge [2]. Therefore, there is a need to develop a reliable and sustainable option to manage leachate treatment effectively.

Thermophilic treatment processes have undergone a fair amount of research and development in recent years. Aerobic thermophilic treatment is one of the advanced treatment technologies used for treatment of high-strength wastewaters. It offers several advantages such as rapid biodegradation rates, low sludge yields, rapid inactivation of pathogenic microorganisms and high loading rate, thus reducing the retention time for treatment and capital cost. Although aerobic thermophilic process offers many benefits, the physical, chemical and biological characteristics of this process are so different from the conventional activated sludge process that the knowledge based from conventional operations is unusable [3].

However, thermophilic processes have also their disadvantages. As activated sludge settling properties depend mainly on the ability of microorganisms to form dense and settleable aggregates (flocs), the increased temperature has been found to deteriorate the sludge settling properties [4]. LaPara and Alleman (1999) [3] reviewed thermophilic aerobic wastewater treatment and reported that the thermophilic sludge settling properties have been varying from poor to excellent. With a few exceptions, sludge settleability in thermophilic processes has more commonly been described as poor rather than good. Sludge yield under thermophilic conditions is commonly thought to be lower than it is under mesophilic conditions. Even though the increase in temperature increases microbial growth rates, the higher decay rate and increased need for maintenance energy reduces the net sludge production.

Therefore, one of the major restraints in the aerobic thermophilic process is the poor bacterial flocculation and problems associated with foams. Poor bacterial settling characteristics resulting from dispersed growing microorganisms which make the separation of biomass difficult and thus, limiting the overall treatment efficiency. Looking into this aspect, an aerobic thermophilic membrane bioreactor (MBR), being able to filter wide ranges of biomass sizes, could be an attractive option for treating high strength landfill leachate. MBR systems are examples of an emerging advanced leachate treatment technology. MBR systems are suspended growth activated sludge treatment systems that rely upon the membrane for liquid/solid separation prior to the discharge of the treated leachate. This study was conducted to evaluate the performance of an aerobic thermophilic MBR in leachate treatment, especially with the complexity of leachate and low sludge yield of thermophilic microorganisms.

2. Materials and methods

2.1. Leachate characterization

The landfill leachate used for the treatment was obtained from Pathumthani Landfill Site (PS), Pathumthani and Ram-Indra Transfer Station (RIS), Bangkok, Thailand. The leachate obtained from each of these sites was characterized (see Table 1) and mixed in proportion to achieve a medium-aged landfill leachate composition (see Table 2).

2.2. Experimental set-up

Experiments of aerobic thermophilic MBR as illustrated in Fig. 1 were conducted in a jacketed cylindrical steel reactor. The double walled steel reactor was passed with hot water to maintain the temperature of the MBR at 45°C. The 6 L working volume was maintained by the leveling tank which automatically opens the valve from the feed tank to the bioreactor when the level of the mixed

Table 1
Composition of leachate obtained from Pathumthani landfill site (PS) and Ram Indra transfer station (RIS)

Parameter	Leachate from site PS	Leachate from site RIS
pH	8.4	3.8
Chemical Oxygen Demand (COD)	4300	68,500
Biochemical Oxygen Demand (BOD)	418	55,880
NH_3-N	1934	390
Total Kjeldahl Nitrogen (TKN)	2186	1006
Conductivity	41,000	36,000
TDS	18,900	16,500
Iron	52.7	860
Copper	0.11	0.17
Zinc	2.28	0.15
Cadmium	0.01	26.5
Lead	0.10	0.44

Note: All units are in mg/L, except for pH and conductivity (μS/cm).

Table 2
Composition of mixed leachate during three experimental phases

Parameters	Unit	Concentration		
		Phase I	Phase II	Phase III
pH	–	7.8–8.2	6.8–7.2	6.3–6.6
COD	mg/L	12,000 ± 1000	12,000 ± 1000	12,000 ± 1000
BOD/COD	–	0.39 ± 0.05	0.57 ± 0.05	0.65 ± 0.05
NH_3-N	mg/L	1700	1300	1000
TKN	mg/L	1900	1600	1300

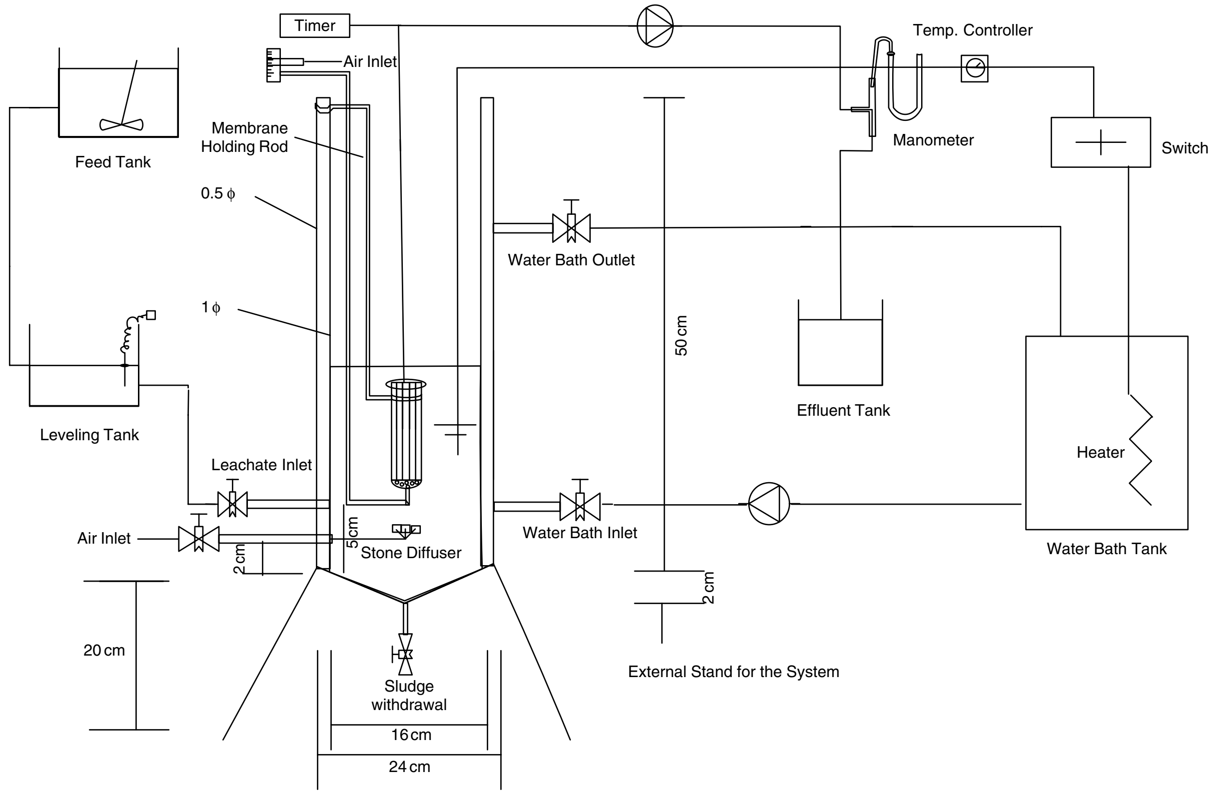

Fig. 1. Experimental set up of the aerobic thermophilic MBR.

liquor has decreased. The reactor was continuously aerated using a stone diffuser placed at the bottom of the reactor. The continuous air supply through the bottom of the membrane provided scrubbing effect in order to minimize biofouling of membrane. An airflow meter was connected to the stone diffuser to maintain the dissolved oxygen concentration in the MBR at around 2–4 mg/L. The pH was maintained at around 6.8–7.0 with the addition of 10% phosphoric acid. Transmembrane pressure (TMP) across the membrane is measured through a U-shaped mercury manometer. Permeate was pumped out with a peristeltic suction pump and a speed controller was used to adjust the flow rate. A ceramic membrane with 22 open fibers with an inner diameter of approximately 2 mm was used. The "outside-in type" ceramic membrane module has a membrane area of 0.04 m^2 and a pure water flux of 411 L/m^2-h-bar. When the membrane was clogged the cleaning was performed by spraying pressurized water to remove the cake layer deposited on the membrane surface. Then the membrane was soaked in 4% sodium hydroxide and 3% sodium hypochlorite mixed solution for 6–12 hrs. Then the membrane was rinsed with water thoroughly to remove the residual chlorine. Acid cleaning was followed by submerging the membrane in 2% hydrochloric acid for 2–15 hrs. The membrane was rinsed again thoroughly with water to remove the residual chemicals. Membrane resistance test was conducted once the cleaning procedure had been completed.

2.3. Operating conditions

The aerobic thermophilic MBR with a working volume of 6 l was operated continuously at 45°C and fed with mixed leachate (with a composition shown in Table 2). The experiments were conducted with different organic loading rates by changing the BOD fraction of the leachate while holding the influent COD value at 12,000 mg/L. The different organic loading rates (OLR) used in this study are presented in Table 3. The mean hydraulic retention time (HRT) was maintained at 24 hrs for all 3 phases of the experiment. Biomass concentration was maintained within the range of 8000–10,000 mg/L of mixed liquor suspended solids (MLSS).

2.4. Analytical methods

Influent and effluent samples were taken randomly for the analysis of COD, BOD, MLSS, TKN, and ammonia. All the analyses were conducted according to the methods given in the *Standard Methods for the examination of water and wastewater [5].*

Sludge characteristics in terms of capillary suction time (CST) and extracellular polymeric substances (EPS) were measured as well. Protein and carbohydrate, being the main components of EPS were analyzed using Lowry method [6] and phenolic sulfuric acid method [7], respectively.

Table 3
Operating conditions of the 3 experimental phases

Phase	Time (days)	BOD/COD	OLR (kg BOD/m^3.d)
1	1–65	0.39	4.9–5.07
2	66–96	0.57	6.27–7.41
3	96–120	0.65	7.15–8.45

3. Results and discussion

3.1. Performance of thermophilic MBR

3.1.1. COD removal Fig. 2 shows the trend of COD removal efficiency in all 3 experimental phases, as differentiated by the BOD/COD ratio and it can be seen that the average COD removal increased from 62 to 79% when the BOD fraction was increased from one phase to another. The average COD removal in phase 1, 2 and 3 were 62, 76 and 79%, respectively. The reason for this increment may be due to the faster rate of biodegradation in thermophilic systems at higher organic loading rates. Similar results have been reported by Rozich and Bordacs (2002) [8] in treating food processing wastewater and by Colvin et al. (2000) [9] and LaPara et al. (2001) [10] in treating pharmaceutical wastewater.

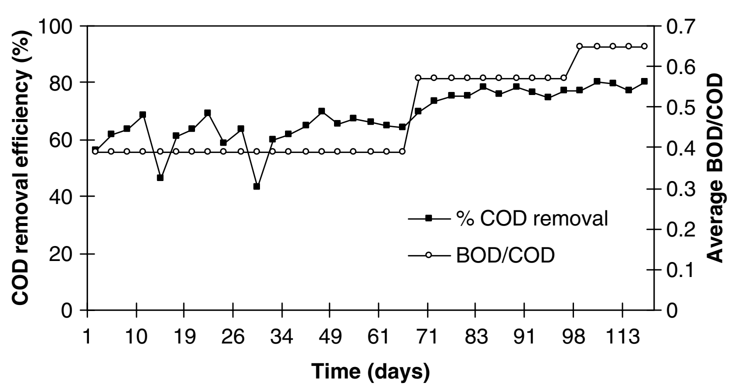

Fig. 2. COD removal efficiency at different BOD/COD ratio.

3.1.2. BOD removal The overall BOD removal efficiency reached more than 97%, which was 20% more than COD removal (Table 4). The removal of BOD started to increase more rapidly as the organic influent was increased in 2nd and 3rd phases. Thus the thermophilic aerobic MBR is efficient in removing biodegradable organic compounds that are found in the complex landfill leachate. However, the low BOD/COD ratio in the effluent indicates that the effluent contains high refractory substances which were either slowly biodegradable organic materials or non-biodegradable materials that were contained in the leachate.

3.1.3. Nitrogen removal Nitrogen removal efficiency in terms of ammonia and TKN were analyzed and shown in Figs. 3 and 4, respectively. It could be seen that the ammonia and TKN removal efficiencies had dropped from 75 to 60% when the ratio of BOD/COD was increased from 0.39 to 0.65. Fig. 5 shows the concentration of ammonia in the influent and effluent (from the MBR). The influent ammonia concentration has declined gradually due to the dilutions made to increase the ratio of BOD/COD. However, the ammonia concentration in the effluent has remained almost in the same range (at 400–450 mg/L) for the entire duration of the experiment. Similar trend was observed with fluctuating TKN analysis at different experimental phases. Lapara and Alleman (1999) [3] has stipulated that ammonia removal phenomenon in thermophilic condition is governed by temperature, mixing and pH,

Table 4
BOD removal at different BOD/COD ratio

BOD (mg/L)		BOD removal (%)	BOD/COD	
Influent	Effluent		Influent	effluent
4714	98	97.9	0.37	0.02
5146	83	98.4	0.40	0.02
7002	97	98.6	0.57	0.03
6944	92	98.6	0.56	0.03
9498	86	99.0	0.65	0.03
8423	78	99.1	0.67	0.03

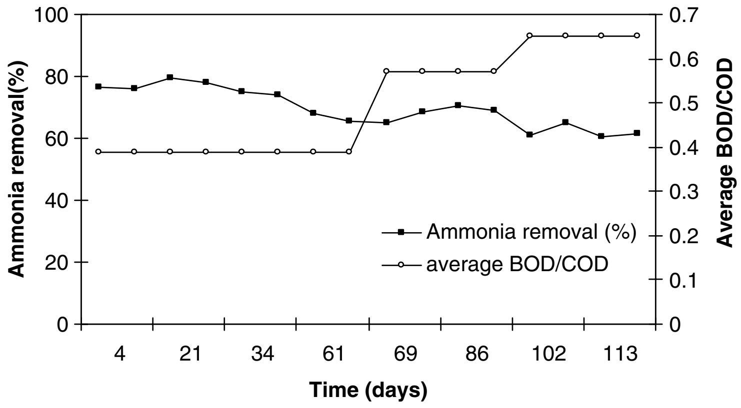

Fig. 3. Ammonia removal efficiency at different BOD/COD ratio.

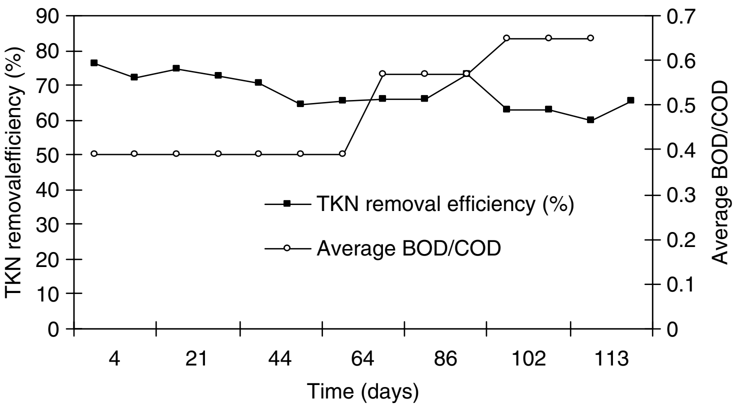

Fig. 4. TKN removal efficiency at different BOD/COD ratio.

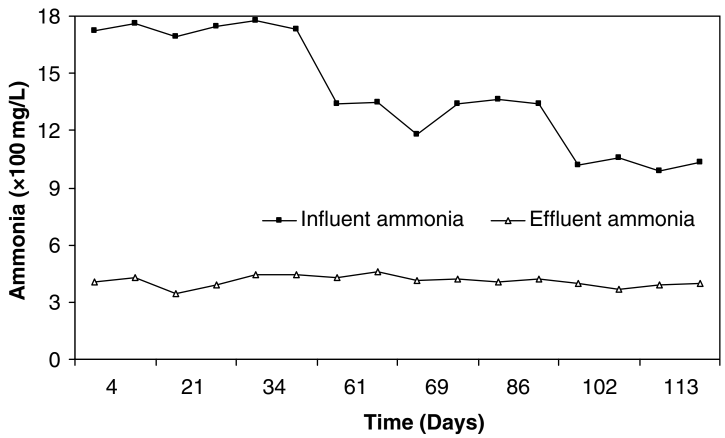

Fig. 5. Variations of influent and effluent ammonia with time

and inhibition of biological nitrification occurs at temperatures greater than 43°C. In this study, it was found that the ammonia was stripped from the wastewaters until the concentrations of 400–450 mg/L regardless of the changes to the BOD/COD ratio and ammonia concentration in the influent.

3.1.4. Extracellular Polymeric Substances (EPS) In MBR systems, membrane biofouling could be affected largely by physicochemical characteristics and the physiology of the sludge as well as the membrane materials. EPS are one of the major causes of membrane biofouling. EPS components could be sub-divided into two parts; bound and soluble. The bound EPS corresponds to the polymeric substances adhered with each other and to the microorganisms, which is related to reversible fouling. At higher bound EPS, stronger cake layer is formed at the surface of the membrane. Soluble EPS indicates the microbial products, which have been produced by the microorganisms and present in the mixed liquor in soluble form. This component is related to irreversible fouling.

Table 5 presents the comparison of EPS concentrations in a thermophilic MBR (present study) and a mesophilic MBR [11]. Soluble and bound EPS concentrations were higher in a thermophilic MBR. However, the ratio of protein to carbohydrate (P/C) is higher in a mesophilic MBR. This supports the literature stating that the thermophilic bacterial populations preferentially utilize

Table 5
Comparison of EPS from thermophilic and mesophilic MBRs

Wastewater	Condition	Soluble EPS (mg/gVSS)	P/C (soluble)	Bound EPS (mg/gVSS)	P/C (bound)	Reference
Leachate	Thermophilic	287.6	0.81	146.4	0.81	This study
Leachate	Mesophilic	119.7	1.66	61.7	1.35	Wichitsathian, 2004

Note: VSS – volatile Suspended Solids; P/C – Protein:Carbohydrate

proteinaceous material as carbon source [12].

3.2. Membrane fouling

The activities that contribute to membrane fouling are varied. It includes adhesion of the colloidal matters and macromolecules on the external and internal surface; growth and adhesion of biofilm on the membrane surface; precipitation of dissolved matters; and age of the membrane [13]. Because of its complex and diverse relationships, fouling definition has not been clearly developed. The main adverse effects of the membrane fouling is the reduction of the permeate flux. In the present study, a constant flux was maintained in the thermophilic MBR. The increase in transmembrane pressure (TMP) in the bioreactor system was measured to observe the rate of fouling of membrane. The membrane was cleaned whenever the TMP reached closer to 60 kPa.

Fig. 6 shows the TMP across the membrane during all three phases. It was observed that the membrane has clogged more frequently than in a mesophilic MBR treating landfill leachate [11]. The probable reason of frequent membrane fouling could be the high EPS formation at thermophilic conditions. The comparative data shown in Table 5 suggests that the amount of EPS produced is almost 2.5 times higher than that observed under mesophilic condition. Literature also suggests that thermophilic sludge contains significantly higher number of small particles compared to mesophilic sludge. In a study conducted by Vogelaar et al. (2002a, 2002b) [14,15], the volume percentage of sludge particles with a diameter of less than 5 μm was 16% for thermophilic sludge and only 4% for mesophilic sludge.

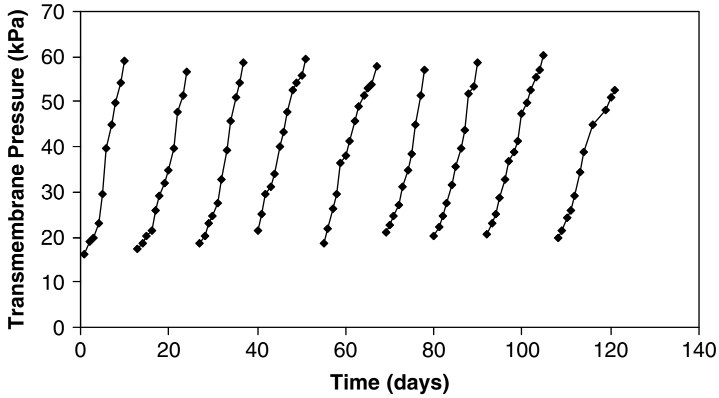

Fig. 6. Variations in transmemebrane pressure (TMP) with time.

4. Conclusion

In this study, the treatment of landfill leachate using aerobic thermophilic MBR has been investigated. The COD removal efficiency was found to increase from 62 to 79% with increased in BOD fraction from 0.39 to 0.65. The average BOD removal was found to be more than 97%, which could imply an effective organic biodegradation by thermophilic microorganisms. Furthermore, the ammonia removal was observed to be decreasing from 75 to 60% with decreasing influent ammonia concentrations. However, it is interesting to note that the ammonia in the effluent was maintained at around 400–450 mg/L regardless of the changes in the influent concentrations. Similar trend was noticed in the TKN results. Thermophilic bacterial population tends to consume proteinaceous compounds than carbohydrate giving lower protein to carbohydrate ratio and higher EPS contents. Higher EPS leads to frequent membrane fouling in thermophilic MBRs than in mesophilic MBRs.

References

[1] J.D. Keenan, R.L. Steiner and A.A. Fungaroli, Landfill leachate treatment. J. Water Pollut. Control Fed., 56(1) (1984) 33–39.

[2] G.J. Farquhar, Leachate production and characteristics. Can. J. Civ. Eng., 16 (1989) 317–325.

[3] T. LaPara and J. Alleman, Review paper; thermophilic aerobic biological wastewater treatment. Water Res., 33 (1999) 895–908.

[4] C. Krishna and M. van Loosdrecht, Effect of temperature on storage polymers and activated sludge settleability. Water Res., 33 (1999) 2374–2382.

[5] APHA, AWWA, WPCF, Standard Methods for the Examination of Water and Wastewater, 20th ed., Washington DC, USA, 1998, ISBN 0–87553–235–7.

[6] O.H. Lowry, N.J. Rosebrough, A.L. Farr and R.J. Randall, Protein measurement with the Folin-Phenol reagents. J. Biol. Chem., 193 (1951) 265–275.

[7] M. Dubois, K.A. Gilles, J.K. Hamilton, R.A. Rebers and F. Smith, Colorimetric method for the determination of sugars and related substances. Anal. Chem., 28 (1956) 350.

[8] A. Rozich and K. Bordacs, Use of thermophilic biological aerobic technology for industrial wastewater treatment. Water Sci. Technol., 46(4–5) (2002) 83–89.

[9] R.J. Colvin, A.F. Rozich, S. Gerber and D. Strom, Thermophilic aerobic treatment of high strength organic production wastes and waste-activated sludge from a pharmaceutical manufacturer: Bench scale test results and full scale design issues, WEFTEC (2000), 73rd annual conference, Anaheim, CA, USA, 2000.

[10] T. LaPara, C. Nakatsu, L. Pantea and J. Alleman, Aerobic biological treatment of a pharmaceutical wastewater: effect of temperature on COD removal and bacterial community development. Water Res., 35 (2001) 4417–4425.

[11] B. Wichitsathian, Application of membrane bioreactor systems for landfill leachate treatment. AIT Dissertation no. EV-04–3, Asian Institute of Technology, Thailand, 2004.

[12] T. LaPara, C. Nakatsu, L. Pantea and J. Alleman, Phylogenetic analysis of bacterial communities in mesophilic and thermophilic bioreactors treating pharmaceutical wastewater. Appl. Environ. Microbiol., 66 (2000) 3951–3959.

[13] B. Gunder, The Membrane Coupled Activated Sludge Process in Municipal Wastewater Treatment, Technomic Publishing Co., Inc., USA, 2001, ISBN 1–56–676959–0.

[14] J. Vogelaar, E. Bouwhuis, A. Klapwijk, H. Spanjers and J. van Lier, Mesophilic and thermophilic activated sludge post treatment of paper mill process water. Water Res., 36 (2002a) 1869–1879.

[15] J. Vogelaar, J. van Lier, A. Klapwijk, M. de Vries and G. Lettinga, Assessment of effluent turbidity in mesophilic and thermophilic activated sludge reactors – origin of effluent colloidal material. Appl. Microbiol. Biotechnol., 59 (2002b) 105–111.