

# **Laboratory Biological Process for Treatment of Leachates from Initial Phases of Landfilling of Mechanically Sorted Organic Residue and Mechanically-Biologically Treated Municipal Solid Waste.**

**Prasad Kaparaju<sup>1\*</sup> and Jukka Rintala**

Dept. of Biological and Environmental Sciences, University of Jyväskylä, Jyväskylä, Finland

## **Abstract**

In this study the combined anaerobic-aerobic treatment of leachates from the initial phases of landfilling of mechanical sorted organic residues and mechanically biologically pretreated municipal solid waste was investigated at 20°C. Anaerobic treatment in UASB followed by aerobic post-treatment in SHARON reactor provided COD removals of about 60-95%. High concentration of nitrite in SHARON reactor and nitrate in ANAMMOX reactor indicated that both processes were either established or in the process of establishment. N removal in combined SHARON and ANAMMOX process was 93-94%. Thus, the combined anaerobic-aerobic treatment showed good potential for COD and N removals. However, the studied leachates were from the initial phase of landfilling and from well isolated test cells, and thus leachates with more strength could be expected on a longer run.

## **Keywords**

Aerobic; anaerobic; ANAMMOX; landfill leachates; MBT; MSOR; MSW; SHARON; UASB;

## **1 Introduction**

Municipal solid waste (MSW) management is experiencing rapid changes in order to promote more sustainable development and to reduce global (e.g. greenhouse gases) and local (leachates) emissions. For many decades, the main practice of MSW management has been – and in many countries still is - landfilling of wastes. However, introduction of source separation of biowaste and its separate stabilisation in 1990's has resulted in decreased emission potential from landfills. Further in the recent years, mechanical and mechanical-biological pretreatment (MBT) of MSW (or MSW after source separation of biowaste) has been also initiated in some countries. In future, MBT and incineration of municipal waste, practised more commonly in Austria and Germany, will become statutory in other countries to operate landfills in an environmentally sustainable manner (Robinson et al., 2005). The introduction of these treatments has resulted and will result in remarkable changes in wastes to be landfilled, and subsequently also the emission potential from landfills. Because of these changes and high variety of the

landfilled wastes, also the characteristics of the landfill leachates vary from different landfills (ROBINSON ET AL., 2005).

Several researchers have studied comprehensively the effect of MBT (LEIKAM AND STEGMANN, 1997; SCHEELHASE AND BIDLINGMAIER, 1997) on MSW and the impacts of MBT on landfill leachate quality (ROBINSON ET AL., 2005). Many detailed studies have shown that very strong leachates that will persist for long periods are generated when untreated mechanically sorted organic residues (MSOR, also termed "residual wastes") are landfilled (WOELDERS AND OONK, 1999; LEIKAM AND STEGMANN, 1999). The high organic content, high moisture content, and small particle size of the MSOR appears to give rise to much higher landfill gas production rates and stronger leachates (ROBINSON ET AL., 2005) leading to a negative environmental impact. On the contrary, an effective MBT process can significantly reduce concentrations of trace organics and of ammoniacal-nitrogen in leachates and can also avoid the peak acetogenic phase of decomposition to produce leachates similar to, or weaker than those from conventional landfills (LEIKAM AND STEGMANN, 1999). However, to meet the statutory requirements prior to discharging MBT landfill leachate to a water course, a secondary treatment for chemical oxygen demand (COD) and nitrogen (N) removal is necessary. In the present study, biological treatment of landfill leachate was investigated using laboratory scale system consisting of upflow anaerobic sludge blanket (UASB) process followed by the combination of SHARON, (Single reactor system for High activity Ammonia Removal Over Nitrogen, HELLINGA ET AL., 1998) and ANAMMOX (anoxic ammonium oxidation) processes, MULDER ET AL., 1992) in a combined SHARON-ANAMMOX concept (VAN LOOSDRECHT AND JETTEN, 1998). The UASB process aimed to remove major COD load while the SHARON and ANAMMOX processes were aimed to remove the nitrogen load. However, the operation of the latter two processes was relative short and thus they can be considered just very preliminary experiments.

## **2 Methodology**

### **2.1 Origin of leachates and leachate characteristics**

Leachates were collected from two separate 100 m<sup>3</sup> landfill cells filled with MSOR and MBT wastes established in Mustankorkea landfill (Jyväskylä, Finland). These cells were constructed in another project and were considered to present leachates produced in the beginning of operation of potential future landfills in Finland. The MSOR represents the waste stream that is currently being landfilled in Finland (except that the waste is not mechanically processed), whereas MBT waste would represent the waste that will be landfilled by the end of this decade in case MBT is introduced. The leachates used represented leachates produced during the initial phases of landfilling and were not avail-

able from any actual landfill during the project. Characteristics of leachates are summarised in Table 1. For the COD removal studies, MSOR leachate was diluted with distilled water in ratio 1:10.

**Table 1** Mean characteristics of leachates used in this study (n=5)

Parameter	MBT	MSOR
pH	7.1	6.0
Total alkalinity (mmol/l)	14	35
TSS (%)	0.75	4.6
VSS (%)	0.73	3.2
BOD <sub>7</sub> (mg/l)	1800	48900
TCOD (mg/l)	4800	76700
SCOD (mg/l)	4500	73400
TKN (mg/l)	385	1830
NH <sub>4</sub> -N (mg/l)	260	960
NO <sub>2</sub> -N (mg/l)	1.7	2.1
NO <sub>3</sub> -N (mg/l)	6.5	19.9
NH <sub>3</sub> -N/NO <sub>2</sub> -N ratio calculated	153	457

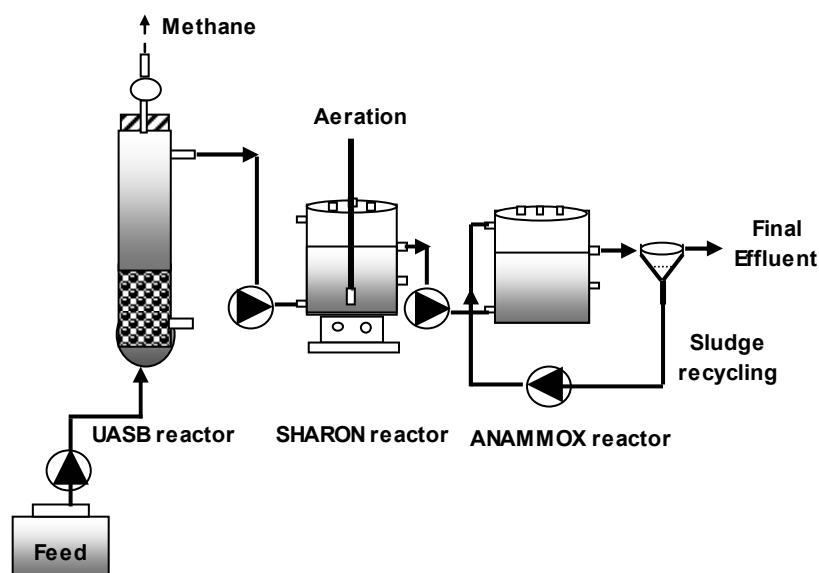
## 2.2 Experimental set-up

The experimental set-up is shown in Fig. 1. All reactors were connected in series so that the effluent from one reactor were drawn and pumped to the next reactor using peristaltic pumps.

### 2.2.1 Anaerobic treatment

As first step, anaerobic treatment of MSOR and MBT leachates was performed in two laboratory UASB glass reactors (inner diameter 5 cm and height 25 cm) of 0.5 l working volume at 20±1°C. UASB reactors were inoculated with 200 ml of granular sludge (6.6 g volatile suspended solids, VSS) originating from an Internal Circulating full-scale reactor

treating industrial potato wastewaters (Finland). Continuous feeding was started immediately. Leachate stored in a feed container at 4°C, was pumped continuously into the bottom of reactors via a peristaltic pump (MasterFlex). Organic loading rates (OLRs) were ranged from 3-4 kg COD/m<sup>3</sup>d for MBT reactor and 5-7.5 COD/m<sup>3</sup>d for MSOR reactor (hydraulic retention time (HRT) of 1 d).



**Figure 1** Experimental set-up.

### 2.2.2 Combined SHARON-ANAMMOX Process

Partial nitrification of N in the effluents from UASB reactors was carried out in two SHARON reactors (3.5 l plexi-glass continuously stirring tank reactors, CSTR) with 2 l working volume and without any biomass retention at 20±1°C. The retention time in SHARON reactors was limited to 4 d. Aquarium air pump (Rena air 100, USA) was used to maintain dissolved oxygen (DO) rate of 1 mg O<sub>2</sub>/l. The reactors were mounted on mechanical stirrers and stirred by magnets continuously at 100 rpm. No pH adjustment was carried out to influence the ammonium/nitrite ratio.

Denitrification of nitrite to dinitrogen gas (N<sub>2</sub>) with ammonium as electron donor was carried out in two ANAMMOX reactors (3.5 l plexi glass CSTRs) at 20±1°C under anoxic conditions. ANAMMOX reactors were operated with a working volume of 1 l and HRTs of 2 d. This was achieved by holding the SHARON effluents in 2 l glass bottles prior to feeding to ANAMMOX reactors. A settler provided at the outlet of the ANAMMOX reactors was used to recycle the settled sludge. No extra addition of base or carbon source to meet the COD requirement e.g. methanol was needed as 50% of the ammonium was needed to convert to nitrite. Moreover, the effluents from AD process contained enough alkalinity (in the form of bicarbonate) to compensate for the acid production if only 50% of the ammonium was oxidized. The reactors were mounted on mechanical stirrers and

stirred by magnets continuously at 75 rpm. Care was taken to avoid entry of air bubbles from SHARON reactors to ANAMMOX reactors

Both SHARON and ANAMMOX reactors were started with 1 l of sludge (21 g VSS) from local sewage treatment plant (Jyväskylä, Finland).

## 2.3 Analyses

pH (Metrohm 744 pH meter) and DO (YSI 550 DO meter) were monitored on daily basis. Pump flow rates were calibrated and adjusted if necessary on every weekday. COD (total, TCOD and soluble, SCOD), biological oxygen demand (BOD<sub>7</sub>), total kjeldahl nitrogen (TKN) and total alkalinity (TA) were determined according to Finnish Standards (SFS-EN 12457-4, Finnish Standards Association, 1998). Ammonium (NH<sub>4</sub>-N), nitrite (NO<sub>2</sub>-N) and nitrate (NO<sub>3</sub>-N) nitrogen were analysed calorimetrically using LANGE LCC 100 (Germany). Total solids (TS), volatile solids (VS), suspended solids (total, TSS and volatile, VSS) were determined according to Standard Methods (APHA, 1998). TKN was analysed using a Kjeltec System 1002 Distilling unit (Perstorp Analytical/Tecator AB, 1995). Methane content in biogas and N<sub>2</sub> were analysed using Gas Chromatography (PerkinElmer).

## 3 Results and Discussion

### 3.1 Characteristics of leachates

The characteristics of MSOR and MBT leachates are presented in Table 1. The low pH of 6 and initial COD of 76,700 mg/l for MSOR leachate suggests that the strong organic leachate was produced during the active acetogenic phase. Similar values in excess of 60,000 mg/l of COD were reported from a 2 year landfill receiving MSOR (ROBINSON ET AL., 2005). On the other hand, COD value of 4800 mg/l with relatively stable pH of 7 suggests that acetogenic phase in MBT cell was absent. A much more significant benefit of pre-treatment was apparent when concentrations of TKN were considered. The TKN in MSOR was about 1830 mg/l whereas this value was below 385 mg/l for MBT leachate. These results were in accordance to the values reported from a 250 days landfill test cells receiving MSOR and MBT wastes (ROBINSON ET AL., 2005). Nevertheless, the BOD/COD ratio of 0.64 for MSOR leachates in the present study was typical for a young landfill leachate. Previously, BOD/COD ratios of 0.5-0.67 were reported for leachates collected from 2 and 3.5 year old municipal landfill sites in Turkey (Timur et al., 2000). The stronger leachates from MSOR than MBT landfill cell could be attributed to the high organic content, high moisture content and small particle size of the MSOR waste.

**Table 2** Mean composition of feed and effluents from the combined anaerobic (UASB) and aerobic process (SHARON/ANAMMOX process) of MBT and MSOR leachates at 20°C (Day 63).

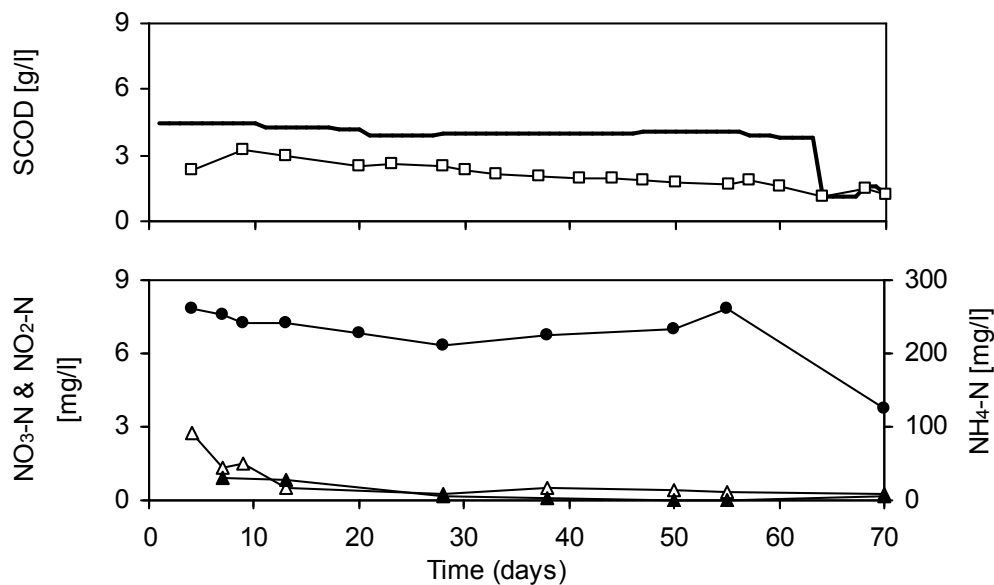
Parameter	MBT				MSOR			
	Feed	UASB effluent	SHARON effluent	ANAMMOX effluent	Feed	UASB effluent	SHARON effluent	ANAMMOX effluent
pH	7.1	7.5	8.2	8.0	6.1	6.8	7.2	7.6
TCOD (mg/l)	2500	2200	2000	1700	8000	1300	1600	710
SCOD (mg/l)	2400	1700	1400	1400	7000	830	80	10
COD removal (%)	--	29.2	41.7	41.7	--	88.1	98.9	99.8
NH <sub>4</sub> -N (mg/l)	2.2	304	2.9	0.3	1.25	167	0	0.09
NO <sub>2</sub> -N (mg/l)	0	1.1	156	0.1	0	4.7	35.5	0
NO <sub>3</sub> -N (mg/l)	6.5	4.6	43	18.5	1.23	0	14.6	9.4
NH <sub>3</sub> -N/NO <sub>2</sub> -N ratio calculated	--	--	0.02	3	--	--	0	--
Total N removal (%)	--	--	33.6†	92.6‡	--	--	70†	93.6‡

†Calculated as:  $\{1 - ([NO_3-N]_{eff} + [NH_4-N]_{eff} + [NO_2-N]_{eff}) / ([NH_4-N]_{in})\} * 100$ . ‡Calculated as:  $\{1 - ([NO_3-N]_{eff} + [NO_2-N]_{eff}) / ([NH_4-N]_{in} - [NH_3-N]_{eff})\} * 100$  and based on highest NH<sub>4</sub>-N value of 252 and 146.5 mg/l in SHARON effluents of MBT and MSOR respectively.

### 3.2 COD removal

The performance of UASB reactors treating MSOR and MBT leachates is presented in Table 2 and Figures 2 and 3. Results showed that anaerobic processes had good potential for COD removal from both MSOR and MBT leachates. At OLRs of 3-5 kg COD/m<sup>3</sup>d and HRT of 1 d in UASB reactors, the COD removals increased along the experimental run. At the end of 70 d of run, COD removals were about 65-80 and 55-97% for MSOR and MBT leachate, respectively. Similar COD removals of 75-91% were reported from UASB reactor treating raw leachates collected from conventional municipi-

pal landfill at 20°C (KALYUZHNYI ET AL., 2003). Subsequently, aerobic (SHARON) post-treatment of the UASB effluents showed to significantly remove the remaining COD from MSOR leachate (>75%), while it was less efficient with MBT leachate (<35%). The combined anaerobic-aerobic treatment provided COD removals of about 80-95% and 60-70% for MSOR and MBT leachate, respectively. The maximum specific methane production ranged between 200 and 240 ml/gCOD for both reactors. These values were somewhat below the theoretically expected value considering the observed COD removal. The low methane production could be attributed to the entrapment of some part of the undigested SS in the reactor sludge bed, consumption of a part of COD for biological sulphate reduction (KALYUZHNYI ET AL., 2003) and also due to supersaturation of psychrophilic effluents by dissolved methane (KALYUZHNYI ET AL., 2001).

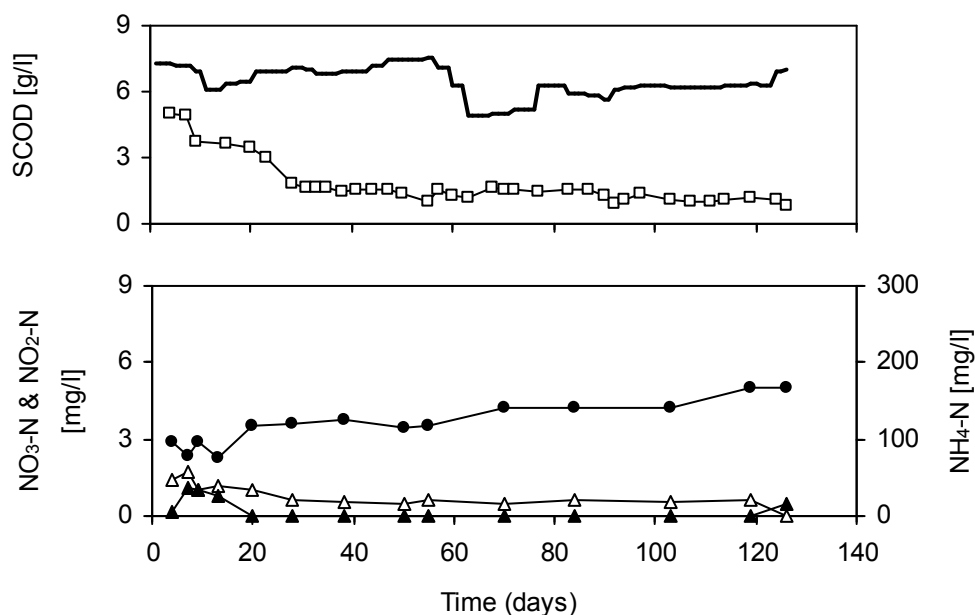


**Figure 2** Performance of UASB reactor treating MBT leachate at 20°C: (a) ■ Load, ○ specific methane production, (b) soluble COD (— influent, □ effluent) and (c) ▲ nitrite-nitrogen ( $\text{NO}_2\text{-N}$ ),  $\Delta$  nitrate-nitrogen ( $\text{NO}_3\text{-N}$ ) and ● ammonia-nitrogen ( $\text{NH}_3\text{-N}$ ) concentration in the effluent.

### 3.3 N removal

Results on N removal using combined SHARON and ANAMMOX process showed an increased ammonium conversion only after 40 d (data not shown). At the end of 70 day run, SHARON reactor seems to an appropriate reactor as it converted much of the ammonium load from UASB effluent into nitrite without applying any pH correction. The concentration of nitrite was higher than nitrate in SHARON reactor, while that of nitrate was higher than nitrite in ANAMMOX reactor (Table 2), indicating that both processes were either established or in the process of establishment. On comparison, ammonium conversion to nitrite in SHARON reactor was much more efficient with MBT (51%) than

with MSOR (21%) leachate apparently due to high levels of N in MBT leachate. Based on the highest ammonium concentrations, 93-94% of total N was removed at the end of the combined SHARON-ANAMMOX process. No ammonium nitrogen was detected while high concentration of  $N_2$  gas was detected in the headspace of ANAMMOX reactors.



**Figure 3** Performance of UASB reactor treating MSOR leachate at 20°C: (a) ■ Load, ○ specific methane production, (b) soluble COD (— influent, □ effluent) and (c) ▲ nitrite-nitrogen ( $NO_2-N$ ), △ nitrate-nitrogen ( $NO_3-N$ ) and ● ammonia-nitrogen ( $NH_3-N$ ) concentration in the effluent.

## 4 Summary

The combined anaerobic-aerobic treatment resulted in 60-90% COD and 93-94% N removals from the initial phase of landfill receiving MSOR or MBT wastes. Anaerobic treatment alone in UASB reactors was quite efficient in removing COD, but aerobic post-treatment contributed also for COD removal. With MBT leachate, COD removals were quite low in aerobic phase. High concentrations of nitrite in SHARON reactor and nitrate in ANAMMOX reactor indicated that both processes were either established or in the process of establishment. However, it must be noted that the studied MSOR and MBT leachates were from the initial phases of landfilling these wastes and from well isolated test cells, and thus the leachates were more strength than the leachates will be on longer run.



## 5 Acknowledgements

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### Author's current address<sup>1</sup>

Asst. Prof. Dr. Prasad Kaparaju  
Institute of Environment & Resources  
Technical University of Denmark  
Building 113, Kgs. Lyngby, DK-2800  
Denmark  
Telephone +45 45 25 15 65

Email :plk@er.dtu.dk

Website: www.er.dtu.dk