

Anaerobic treatment of Hong Kong leachate followed by chemical oxidation

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Abstract Landfill leachate of Hong Kong was first treated by the upflow anaerobic sludge blanket (UASB) process. At 37 °C, pH 7.1–8.5 and a HRT of 5.1–6.6 days, the process removed 66–90% of COD in the leachate for loading rates of 1–2.4 g-COD/l day depending on the strength of landfill leachate. The final effluent contained 1440–1910 mg-COD/l and 70–91 mg-BOD/l. About 92.5% of the total COD removed was converted to methane and the rest was converted to biomass with an average net growth yield of 0.053 g-VSS/g-COD-removed. The granules developed in the UASB reactor were 0.5–1.5 mm in size and exhibited good settleability. The UASB effluent was then further polished by two oxidation processes. The UASB-ozonation process removed 93.0% of the 12900 mg/l of COD from the raw leachate. The UASB-Fenton-ozonation process improved the COD removal efficiency to 99.3%. The final effluent had only 85 mg/l of COD and 10 mg/l of BOD₅. Ozonation was most effectively conducted at pH 7–8 with the addition of 300 mg/l of H₂O₂ and for the duration of 30 min. Ozonation also significantly improved the biodegradability of the organic residues. Nearly 50% of these residues could be used as carbon source in denitrification.

Keywords Anaerobic treatment; Fenton reaction; leachate; ozonation

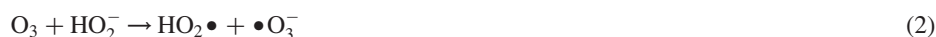
Introduction

The organic residues in landfill leachate are in general recalcitrant to biodegradation in traditional wastewater treatment facilities, and some are also colloidal in nature. As a result, biological treatment of leachate is often not effective. Hong Kong produces about 9500 tonnes of municipal solids waste daily. They are collected and disposed of at three landfill sites with ages varying from seven to nine years. The leachate is at present treated in oxidation ponds on site with a hydraulic retention time (HRT) of 30 days (Chen, *et al.*, 1997). The process is costly because of its strong energy demand. It is, thus, warranted to develop a more cost-effective alternative.

This project was conducted to study the feasibility of treating Hong Kong leachate with anaerobic process, replacing the existing extended aeration, followed by chemical oxidation. Anaerobic process has become a mature wastewater treatment technology in the past decade. The process not only saves aeration energy but also produces methane, which is a readily useful fuel. In addition, it produces only about 10% of waste sludge as compared to aerobic processes. Thousands of full-scale treatment plants have been installed worldwide (Fang and Liu, 2001). However, due to the recalcitrant nature of the organic residues in the leachate, anaerobic treatment alone was not sufficient. The anaerobic effluent was further treated in this study with a modified ozonation process.

Ozonation is a widely accepted technology used for the final disinfection of water and wastewater. In recent years, it has also been increasingly used for the treatment of refractory and toxic wastewaters from various industries, such as tannery (Jochimsen *et al.*, 1997), pulp and paper (Zhou and Smith, 1997) and pharmaceutical (Hofl *et al.*, 1997), as

well as landfill leachate (Primi *et al.*, 1997). The oxidation potential of ozone lies on its conversion into the free radical $\bullet\text{OH}$ (Langlais *et al.*, 1991) through a series of chain reactions. The free radical $\bullet\text{OH}$ is of high oxidation potential ($E^\circ = 2.80\text{ V}$) and capable of reacting with many organic species within a millisecond (Peyton, 1988). However, it has been found that the presence of H_2O_2 considerably enhances the formation of $\bullet\text{OH}$. H_2O_2 could be dissociated into HO_2^- (see Reaction 1), which could further react with ozone forming two $\bullet\text{OH}$ precursors, $\text{HO}_2\bullet$ and $\bullet\text{O}_3^-$ (see Reaction 2) (Gulyas, 1997):



It is thus advantageous to conduct ozonation with a small dosage of H_2O_2 in the treatment of wastewater (Gulyas *et al.*, 1995) and drinking water (Hirvonen *et al.*, 1996). In this study ozonation of anaerobic effluent was accompanied by dosage of H_2O_2 . Furthermore, a fraction of anaerobic effluent was partially oxidized using the Fenton coagulation process (Lau *et al.* 2001) prior to ozonation for comparison.

Materials and methods

UASB treatment

A 2.8 litre UASB (upflow anaerobic sludge blanket) reactor with an internal diameter of 84 mm and a height of 500 mm (Fang *et al.*, 1995) was used in this study. On top of the reactor was a gas-liquid-solid separator with an internal diameter of 114 mm and a height of 250 mm, making a filled volume of 2.0l. Volumetric loadings were calculated based on the reactor volume alone, excluding the volume of the separator. The reactor was water-jacketed and operated at a constant temperature of 37 °C for 354 days. A variable-speed peristaltic pump (Cole-Parmer, Masterflex) was used to feed raw leachate into the reactor, forcing an equal volume of supernatant out of the effluent line. The raw leachate was collected from a Hong Kong landfill site and was kept in the refrigerator at 4 °C. The reactor was seeded with 1 litre each of UASB sludge (26.7 g-VSS/l) and settled activated sludge from the oxidation pond (10.8 g-VSS/l) at the landfill site. The UASB seed sludge was from previous studies treating wastewater containing proteinaceous and carbohydrate pollutants.

Ozonation

The UASB effluent during days 150–195 was sampled for ozonation treatment. The average effluent had 1440 mg/l of COD and 81 mg/l of BOD_5 . Ozone was generated from oxygen using an Osmonics ozonator (System V5-0-813). The oxygen was fed into the ozonator at 140 kPa and the voltage was controlled at 90 V. The ozonator produced a gas stream containing 52.2 mg/l of ozone. The gas bubbled through a diffuser into 500 ml of effluent in a 2000 ml reactor at room temperature. The flow rate of the ozonator gas stream was maintained at 2400 ml/min. A fraction of UASB effluent was partially oxidized by the Fenton process at pH 5 with 300 mg/l of Fe^{2+} and 200 mg/l of H_2O_2 , as described previously (Lau *et al.*, 2001).

Analyses

The measurements of COD, BOD_5 (5-day biochemical oxygen demand), TKN (Total Kjeldahl Nitrogen) and $\text{NH}_3\text{-N}$ followed the procedures of *Standard Methods* (APHA, 1985). The measurements of VFA followed those reported previously (Fang *et al.* 1995). Nitrate was analyzed by an ion chromatograph (LC-10, Shimadzu) equipped with an Allsep™ anion column.

Results and discussion

UASB treatment of raw leachate

Raw leachate containing 4750–15700 mg-COD/l was treated in the UASB reactor. Throughout the experiment, the pH of mixed liquor in the reactor was kept at pH 7.1–8.5, which was mainly due to the high alkalinity buffer in raw leachate. The 354 day experimental program was divided into two stages: Stage 1 was the startup operated at different organic loading rates ranging 0.7–5.6 g-COD/l day so that the optimum organic loading rate could be determined and the seed sludge could be gradually transformed into leachate-degrading granules; Stage 2 was operated by feeding the UASB reactor with leachate collected from the landfill site at different time. Figure 1 illustrates operational conditions and performance of the UASB reactor in all phases throughout this study, including (a) the COD loading rate, (b) influent COD, (c) HRT, and (d) efficiency of COD removal.

Organic removal during startup phase

The UASB reactor was operated at an organic loading rate of 0.9 g-COD/l day with 5200 mg/l of COD in the leachate. The efficiency of COD removal increased from the initially observed 65% to 75% in two weeks. The loading was then increased to 2.4 g-COD/l day for one more week and then on Day 26 to 5.7 g-COD/l day. When the loading was increased to 5.7 g-COD/l day, the efficiency of COD removal was immediately reduced to 47%. The reactor slightly recovered when the organic loading rate was reduced to 2.5 g-COD/l day (Day 36), but turned worse again when the loading was increased on Day 47. From Day 68 to Day 120, an organic loading rate of 0.7 g-COD/l day stabilized the granulation and the reactor performance. The COD removal efficiency was maintained at 60%. A distinct granulated sludge bed was observed starting Day 78. The granules in the reactor were found to be 0.5–1.5 mm in size and exhibited good settleability.

Effect of organic loading rate on UASB process

After the first 120 days of operation, four consecutive batches of raw leachate containing 4750–15700 mg-COD/l and 1310–2260 mg-NH₃-N/l were fed into the UASB reactor. Table 1 summarizes the performance of the UASB reactor and the characteristics of raw leachate and the UASB effluent from Day 121 to Day 354. Although high concentration of ammonia could inhibit the methanogenic activity (Koster and Lettinga, 1984), the UASB process could still remove 66.1–90.4% of COD, and 96.0–98.4% of BOD₅. The UASB effluent having a BOD₅ of only 70–91 mg/l was recalcitrant to further biodegradation. The NH₃-N concentration was increased by 8.4–13% after the UASB treatment

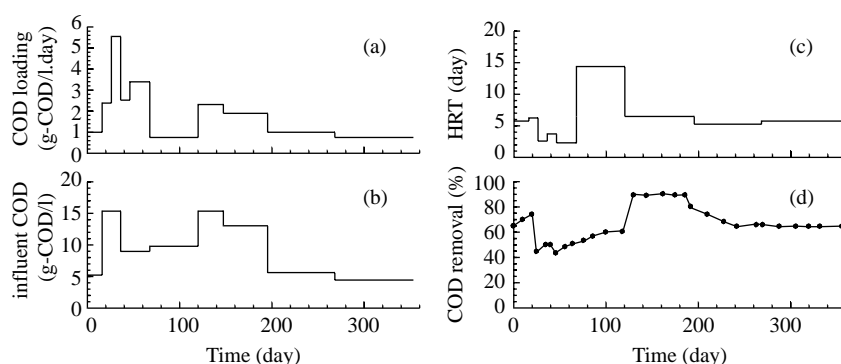


Figure 1 Operational conditions and performance of UASB reactor: (a) COD loading rate, (b) influent COD, (c) HRT, and (d) efficiency of COD removal

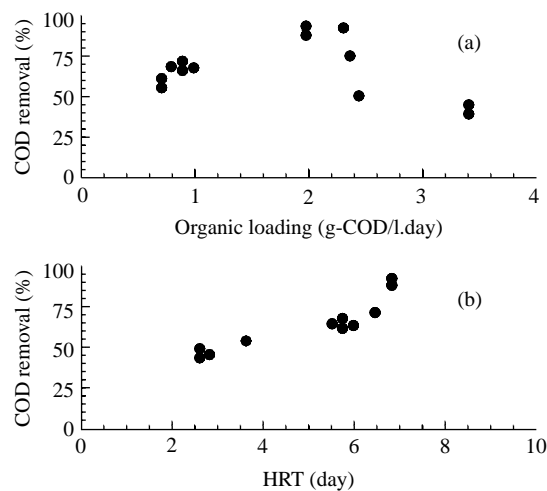
Table 1 Characteristics of raw leachate and UASB effluent

		Day 121–150	Day 151–195	Day 196–268	Day 269–354
Raw leachate	pH	7.7	7.8	7.6	7.9
	COD (mg/l)	15700	12900	5626	4750
	BOD ₅ (mg/l)	4200	5062	3350	2270
	TOC (mg/l)	4600	3240	1185	1190
	NH ₃ -N (mg/l)	2260	2100	1552	1310
UASB effluent	pH	8.5	8.7	8.9	8.9
	COD (mg/l)	1500	1440	1910	1610
	BOD ₅ (mg/l)	75	81	70	91
	TOC (mg/l)	470	370	381	390
	NH ₃ -N (mg/l)	2540	2306	1753	1480
HRT	(day)	6.6	6.6	5.5	6.1
COD loading rate	(g-COD/l day)	2.37	1.95	1.02	0.78
COD removal	(%)	90.4	88.8	66.1	66.1
BOD ₅ removal	(%)	98.2	98.4	97.9	96.0
TOC removal	(%)	89.8	88.6	67.8	67.2
NH ₃ -N increase	(%)	12	8.4	13	13
Biomass accumulated	(g-VSS)	26.3	27.8	28.5	29.1

because of the conversion of organic nitrogen into NH₃-N, which also resulted in an increase of pH.

Fang and Chui (1993) demonstrated that the COD removal efficiency of a UASB reactor was mainly dependent on the COD loading rate, which could be adjusted by varying HRT and/or the COD level in the wastewater. Figures 2a and 2b illustrate the effects of organic loading rate and HRT on the COD removal efficiency. Since there existed certain amount of refractory organics in raw leachate, the COD removal efficiency remained at 60% when the organic loading rate was kept at less than 1 g-COD/l day during the startup process. But it was maintained at 89–91% at the organic loading rate of 1.9–2.4 g-COD/l day. Beyond this loading rate, the performance of the reactor deteriorated sharply (Figure 2a). Figure 2b illustrates that the COD removal efficiency increased with the HRT and attained 91% at an HRT greater than 6.6 days.

Figure 3 illustrates the linear relationship between specific methane production rates and specific substrate utilization rates in the UASB reactors. The slope in Figure 4 indicates that, of all the COD removed, an average of 92.5% was converted to methane, and

**Figure 2** Effects of (a) organic loading and rate and (b) HRT on COD removal efficiency

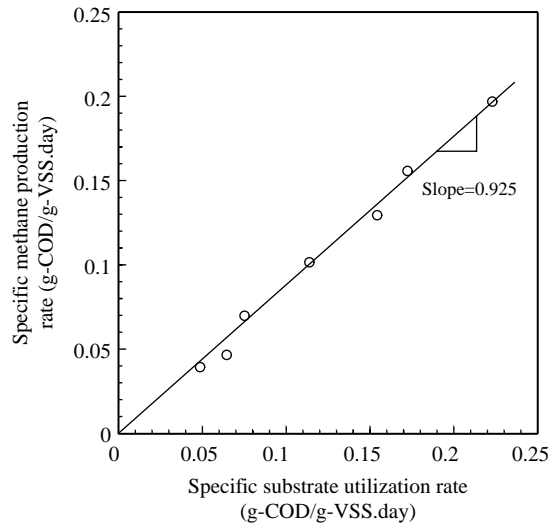


Figure 3 Relation between specific methane production rate and specific substrate utilization rate

the remaining 7.5% was, presumably, utilized for the bacterial growth. Assuming a chemical formula of $C_5H_7O_2N$, biomass has a COD-equivalent of 1.42 g-COD/g-VSS. Thus, the yield for the granular sludge was estimated as 0.053 g-VSS/g-COD-removed ($0.075/1.42 = 0.053$), which was comparable to those of anaerobic sludge treating leachate reported: 0.05–0.09 g-VSS/g-COD by Kennedy *et al.* (1988), 0.041 g-VSS/g-COD by Chang (1989), and 0.071 g-VSS/g-COD by Shin *et al.* (1999).

H_2O_2 dosage in ozonation

A series of ozonation experiments were conducted in parallel. The pH of the UASB effluent treated by Fenton reaction was initially kept at pH 6. The H_2O_2 dosage varied from nil to 500 mg/l. Figure 4 illustrates that the H_2O_2 dosage did not have a noticeable effect on the COD removal in the first 5 minutes. But thereafter the removal efficiency increased not only with time but also with the H_2O_2 dosage. The COD removal efficiency began to level off at about 30 min in all experiments. Without the addition of H_2O_2 , simple ozonation removed 23% of COD; but the efficiency increased to 32% with 300–500 mg/l of H_2O_2 .

Since $\bullet OH$ is highly reactive, the efficiency of oxidation processes often depends on the rate of $\bullet OH$ generation (Hoigne, 1997). Since the addition of H_2O_2 would enhance

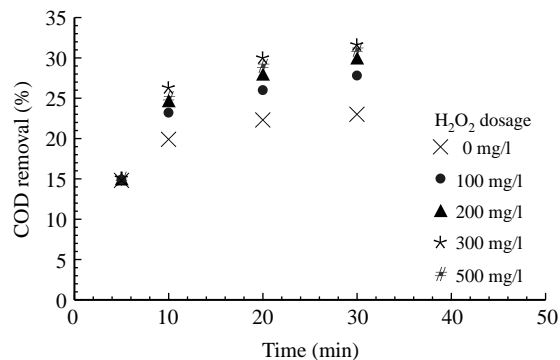


Figure 4 Effect of duration and H_2O_2 dosage in ozonation on COD removal

the production of two free-radical precursors of $\bullet\text{OH}$, one would expect the COD removal efficiency would increase with the $\bullet\text{OH}$ dosage, as in the cases of treating industrial wastewater (Paillard *et al.*, 1988; Gulyas *et al.*, 1995), and contaminated groundwater (Hirvonen *et al.*, 1996). However, Figure 4 illustrates that increasing the H_2O_2 dosage from 300 mg/l to 500 mg/l did not improve the COD removal. This could be due to the following reaction, which shows that H_2O_2 in excess could become an $\bullet\text{OH}$ scavenger (Buxton *et al.*, 1988).



It was also observed that during the first five minutes of ozonation, the gas generation was intense and the gas bubbles produced were finer than those at later stages. This could be due to the presence of surface active chemicals at the initial stage, which were degraded as ozonation proceeded (Pacana and Gurol, 1982). Thus, further increase of H_2O_2 dosage would not improve the COD removal.

Effect of initial pH in ozonation

The effect of pH on ozonation with H_2O_2 addition was unclear. Many reported that high pH is preferred for ozonation (Pacana and Gurol, 1982; Singer and Gurol, 1983), which is consistent with Reaction (1). On the other hand, high pH also favors the formation of CO_3^{2-} and HCO_3^- , both which are scavengers of $\bullet\text{OH}$ (Langlais *et al.*, 1991) as shown in the following reactions, and thus would lower the ozonation efficiency.



In order to clarify the effect of pH on ozonation, a series of experiments were conducted. In these experiments, ozonation was conducted with 300 mg/l of H_2O_2 addition and lasted for 30 minutes. Figure 5 illustrates that the optimal removal efficiency of increased drastically from 32% at pH 6 to 52% at pH 7–8; further increase of pH lowered the efficiency slightly.

Effluent quality of ozonation

Two ozonation experiments were conducted in parallel for comparison, one treating the UASB effluent and the other treating the supernatant of Fenton coagulation. Both experiments were conducted at pH 7 with 500 mg/l of H_2O_2 dosage and lasted for 60 minutes. Results showed that ozonation removed 37% of the COD in the UASB effluent, and 78% of the COD in the supernatant of Fenton coagulation. The latter was considerably higher

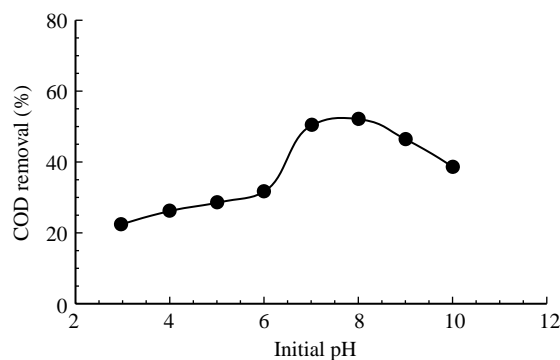


Figure 5 Effect of initial pH on COD removal

than the 52% removal observed in the experiment which ozonation lasted only 30 minutes. The off-gases contained 22.4 mg-O₃/l and 25.3 mg-O₃/l, respectively, and the ozone concentration in both mixed liquors was 25 mg-O₃/l. Therefore, based on mass balance, removing each gram of COD of the refractory organic residues in the UASB effluent consumed 16 g of O₃, and corresponding O₃ consumption for the Fenton coagulation was 25 g. The characteristics of UASB effluent before and after ozonation are listed in Table 2, along with those of the Fenton-coagulation supernatant for comparison.

These results showed that 97.0% of COD of the raw leachate was removed by UASB plus Fenton-coagulation, and further treatment using ozonation would improve the overall COD removal to 99.3%. The final effluent had 85 mg/l of COD and 10 mg/l of BOD₅ after ozonation. On the other hand, ozonation of UASB effluent would only remove 93.0% of COD overall. Table 2 also shows that ozonation resulted in the decrease of TKN and the increase of NH₃-N. A small fraction of organic-N was oxidized to form NO₃⁻-N.

The dosages of Fe²⁺ and H₂O₂ were 300 mg/l and 200 mg/l, respectively. According to a recent study (Lau *et al.*, 2001), H₂O₂ and excess Fe²⁺ synergetically removed organic residues by free-radical oxidation as well as coagulation. In this study, the Fenton coagulation process removed 73% of COD from the UASB effluent, leaving 394 mg/l of COD in the effluent. Under such a condition, removing each gram of COD required 0.29 g of Fe²⁺ and 0.19 g of H₂O₂, which were consistent with results of the previous study (Lau *et al.*, 2001).

Biodegradability of ozonation products

Table 2 shows that the BOD₅/COD ratio increased after ozonation for both the UASB effluent and the Fenton-coagulation supernatant. This indicates that the biodegradability of the residual organics in both streams increased as a result of ozonation. Similar observation had been reported by Anderson *et al.* (1984), and Fazzini and Young (1994). This could be due to the breakdown of complex humic and aromatic substances into smaller and more biodegradable organics. Two series of experiments were conducted using the organic residues in the final effluent of the UASB-Fenton-ozonation process as substrate for denitrification and anaerobic methane production. The biodegradability of the organic residues are measured by the COD removal efficiency during the tests. Results show that the organic residues were more easily degraded for denitrification than for methane production. Both activities, however, increased with the H₂O₂ dosage during ozonation.

Upon ozonation, the total-VFA (mostly acetate and i-caproate) increased from 24 mg-COD/l to 53 mg/l in 10 min without the addition of H₂O₂. This indicates that ozonation could convert some organic residues into VFA; however, further ozonation mineralized the VFA and gradually lowered the VFA content. Also, the formation of

Table 2 Effects of ozonation on the characteristics of UASB effluent and supernatant of Fenton coagulation

	UASB effluent		Supernatant of Fenton coagulation	
	Before	After	Before	After
BOD ₅ (mg/l)	81	103	26	10
COD (mg/l)	1440	905	394	85
BOD ₅ /COD ratio	0.056	0.114	0.066	0.118
TKN (mg/l)	2519	2366	2497	2446
NH ₃ -N (mg/l)	2306	2320	2314	2376
Org-N (mg/l)	213	46	183	70
NO ₃ ⁻ -N (mg/l)	0	25	0	22

VFA decreased with the increased dosage of H_2O_2 due to strong oxidizing effect provided by the added H_2O_2 which also resulted in the mineralization of VFA. With the addition of 300 mg/l of H_2O_2 the total-VFA content was gradually decreased to 15 mg-COD/l after 30 minutes of ozonation. It represented merely about 18% of the organic residues which had 85 mg/l of COD as shown in Table 2. And yet results showed that nearly 50% of the organic residues could be used as substrate for denitrification. This seems to suggest that most of these substrates used for denitrification were not VFA.

Conclusions

Results from this study show that landfill leachate could be effectively treated by the UASB process. At 37 °C, pH 7.1–8.5 and a HRT of 5.1–6.6 days, a laboratory-scale UASB reactor was capable of removing 66–90% of COD for loading rates of 1–2.4 g-COD/l day depending on the strength of landfill leachate. The final effluent contained 1440–1910 mg-COD/l and 70–91 mg-BOD/l. About 92.5% of the total COD removed was converted to methane and the rest was converted to biomass with an average net growth yield of 0.053 g-VSS/g-COD-removed. The granules developed in the UASB reactor were 0.5–1.5 mm in size and exhibited good settleability. The UASB effluent was effectively polished by two oxidation processes. The UASB-ozonation process removed 93.0% of the 12900 mg/l of COD from the raw leachate. The UASB-Fenton-ozonation process improved the COD removal efficiency to 99.3%. The final effluent had only 85 mg/l of COD and 10 mg/l of BOD₅. Ozonation was most effective at pH 7–8 with the addition of 300 mg/l of H_2O_2 and for the duration of 30 min. Ozonation also significantly improved the biodegradability of the organic residues. Nearly 50% of these residues could be used as carbon source in denitrification.

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