

## Treatment of landfill leachates: aerobic bio-oxidation and post-ozonation

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**Abstract.** The purpose of the study was to improve the purification process of landfill leachate. Aerobic bio-oxidation of landfill leachate and post-ozonation of biologically treated leachate were studied. The results of experiments indicate that aerobic bio-oxidation is efficient for purifying young landfill leachate only under stable operating conditions with a long retention time. Subsequent ozonation results in significantly improved biodegradability of the wastewater.

**Key words:** landfill leachate, aerobic bio-oxidation, post-ozonation.

### INTRODUCTION

Sanitary landfills are used for the disposal of municipal refuse. The generated landfill leachate is considered hazardous waste and can cause pollution of the surrounding soil and surface and ground water. Therefore, in order to avoid environmental deterioration, landfill leachate must be collected and treated before being discharged into the environment.

Landfill leachate is generated by excess rainwater percolating through the waste layers. Combined physical, chemical, and biological processes in the waste material transfer pollutants from the waste to the percolating water. Landfill leachate may be characterized as a water-based solution of four groups of pollutants [1]:

– dissolved organic matter (expressed as chemical oxygen demand (COD) or total organic carbon), volatile fatty acids, and more refractory compounds, for example, fulvic-like and humic-like compounds;

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- inorganic macrocomponents: Ca, Mg, Na, K,  $\text{NH}_4^+$ , Fe, Mn,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{HCO}_3^-$ ;
- heavy metals: Cd, Cr, Cu, Pb, Ni, and Zn;
- xenobiotic organic compounds originating from household or industrial chemicals and present in low concentrations in the leachate. These compounds include also a variety of aromatic hydrocarbons, phenols, and chlorinated aliphatics.

Leachate composition varies significantly depending on solid waste characteristics, age of landfill, and climatic conditions [2, 3].

The increased concern about environmental pollution linked to landfill leachate has resulted in many investigations addressing the treatment and purification of highly contaminated streams. The main applicable methods are biological, physicochemical, chemical, and membrane separation processes. Physicochemical processes and chemical oxidation are suitable for treatment of aged landfill leachates, whereas biological processes are more suitable for treatment of young landfill leachates [4]. Among wastewater technologies, biological oxidation is probably one of the most advantageous treatments characterized by low maintenance costs and extensive knowledge of these types of processes, especially aerobic processes. However, for the wastewater containing heavily biodegradable or even toxic compounds like landfill leachate, it is impossible to achieve the needed degree of purification when using only aerobic biological methods [5]. Therefore, a number of innovative treatment methods have been investigated where the aerobic biological treatment is combined with anaerobic biological treatment [3, 5] or with chemical oxidation treatment [4, 6, 7].

The aim of the study was to establish the application possibilities and expected maximum efficiency of conventional aerobic bio-oxidation of landfill leachate and post-ozonation of biologically treated leachate.

## **AEROBIC BIO-OXIDATION: EXPERIMENTAL AND RESULTS**

In the experiments a synthetic solution corresponding by its composition to leachate of young landfills [3] was used as the wastewater. The synthetic leachate was made daily by dilution of concentrated solutions. The organic part of the concentrated synthetic solution had the following composition: acetic acid – 40 g/L, propionic acid – 15 g/L, butyric acid – 15 g/L, valeric acid – 5 g/L, acetone – 5 g/L, ethanol – 5 g/L, propanol – 5 g/L, carbohydrates – 3 g/L, toluene – 1 g/L, phenol – 1 g/L. The main constituents of the inorganic part were Na, Ca, Mg, Fe, K,  $\text{SO}_4^{2-}$ , and  $\text{Cl}^-$ . In addition, it contained Mn, Zn, Cr, Co, Ni, and Cu as impurities. The nutrients N (as  $\text{NH}_4$ ) and P were added proportionally to the organic (destructible) part during the dilution of the synthetic solution. The main parameters of the synthetic solutions used in the experiments were: COD – 2100 to 2800 mg/L;  $\text{BOD}_7$  – 1100 to 1500 mg/L, pH – 6. The composition of the synthetic landfill leachate with COD of 2800 mg/L is given in Table 1.

**Tabel 1.** Composition of the synthetic landfill leachate

Parameter	Value	Parameter	Value
COD, mgO/L	2800	SO <sub>4</sub> <sup>2-</sup> , mg/L	400
BOD <sub>7</sub> , mgO/L	1500	Cl <sup>-</sup> , mg/L	350
pH	6	Na, mg/L	450
Acetic acid, g/L	0.87	Ca, mg/L	500
Propionic acid, g/L	0.32	Mg, mg/L	100
Butyric acid, g/L	0.32	Fe, mg/L	100
Valeric acid, g/L	0.11	K, mg/L	100
Acetone, g/L	0.11	P, mg/L	15
Ethanol, g/L	0.11	Mn, mg/L	10
Propanol, g/L	0.11	Zn, mg/L	0.5
Hydrocarbons, g/L	0.06	Cr, mg/L	0.01
Toluene, g/L	0.02	Co, mg/L	0.01
Phenol, g/L	0.02	Ni, mg/L	0.05
NH <sub>4</sub> <sup>+</sup> , mg/L	250	Cu, mg/L	0.02

In aerobic bio-oxidation the experimental apparatus consisted basically of a laboratory bioreactor. The apparatus was operated in a continuous flow mode and a long-lasting uninterrupted operation was guaranteed by a computer-based automatic control system.

The biological reactor was an aerobic activated sludge reactor with aeration (volume 7.5 L) and settling (volume 2.5 L) chambers. The reactor was supplied with a peristaltic feed pump (Masterflex 7518), sensors for temperature and dissolved oxygen concentration measurement (Marvet Junior), compressors for air transportation (MaximaR), and dispersion diffusers. Aerobic conditions and mixing in the reactor were assured through continuous aeration with submerged diffusers in the aeration unit.

The influent water was led into the reactor by an automatically controlled feed pump. The effluent water flowed out from the bioreactor through an overflow pipe. The transfer of oxygen from air into water was assured with two air compressors, from which one operated continuously and the other was switched on automatically when the concentration of dissolved oxygen had become less than a given value.

Samples from the influent and the effluent were analysed for COD and biochemical oxygen demand (BOD<sub>7</sub>). Biomass concentration and settleability in the bioreactor were also determined. COD was analysed using Hach Standard Method and BOD<sub>7</sub> was analysed according to procedure 5210 of the Standard Methods for the Examination of Water and Wastewater.

Biomass used in the experiments was grown in the aerobic bioreactor. There was no introduction of organisms to the bioreactor through inocula. Organisms were cultured simply through the introduction of untreated synthetic landfill leachate. The influent strength was increased step by step.

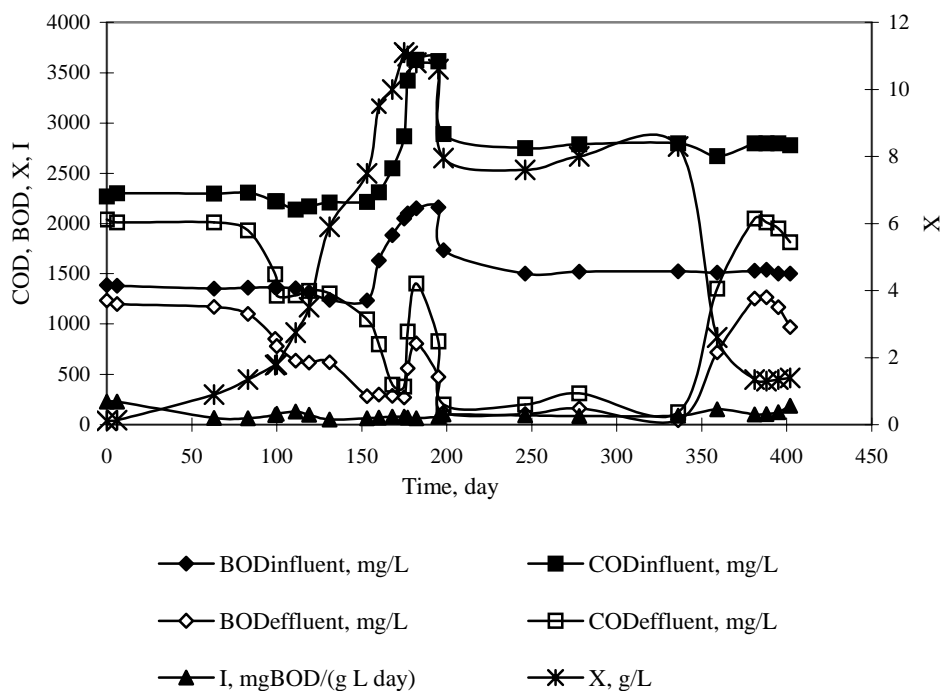
The experiments of aerobic bio-oxidation were conducted in a continuous-flow mode. In the beginning the flow rate of the influent was 1.67 L/day, which

corresponded to retention time of 6 days. Under normal operational conditions the dissolved oxygen concentration was maintained in the range from 2 to 4 mg/L. In this period the biomass became adapted to the experimental conditions.

Figure 1 presents the variation of parameters characterizing aerobic bio-oxidation. Organics removal rate was calculated in units of mgBOD/L removed per g of biomass per day. The operating period beginning from the instant when the performance of the aerobic bioreactor had stabilized was under observation. On day 60, the retention time was reduced to 3 days. In this period the biomass concentration in the bioreactor was relatively low and, consequently, efficiencies of the process both in terms of COD and BOD were low. The efficiency in terms of COD decreased with decreasing retention time and was in the range of 9–33%, and the efficiency in terms of BOD was 12–43%.

From day 110 the reactor operated at a retention time of 2 days. By day 153 the biomass concentration was increased up to 7.5 g/L, and the efficiency in terms of BOD was 75% and in terms of COD 52%. The starting period was considered complete and the planned experiments were started.

First the effect of BOD loading on the COD and BOD removal and biomass growth were studied. Methanol was added to the synthetic landfill leachate in the amounts that increase BOD of the influent to 2100 mg/L. At this loading (days 175–195), the concentration of biomass in the reactor became 10–11 g/L. The efficiency of the process maintained its level, but COD and BOD of the effluent



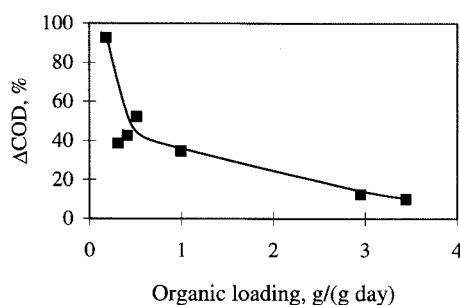
**Fig. 1.** Evolution of the parameters of continuous aerobic bio-oxidation. I, organics removal rate; X, biomass concentration.

increased. On day 195 the adding of methanol was finished and this resulted in a rapid decrease of the effluent COD and the COD removal reached 93%, also BOD removal ranged around 90–93%.

To avoid the so-called dead zones in the bioreactor and insufficient oxygen supply under the conditions of continuous growth of biomass, regular removal of excess biomass from the reactor was started and the biomass concentration was maintained at the level of 8–9 g/L. The bioreactor operated under these conditions during the period from day 196 to 330.

On day 330, the organic loading rate (kgCOD/(kg of biomass per day)) was increased: the concentration of constituents in the synthetic landfill leachate was increased so that COD and BOD of the influent achieved 2800 and 1500 mg/L, respectively. The changes in the loading were followed by disturbances in the performance of the bioreactor (foaming, decrease of settleability, and loss of biomass with effluent). These phenomena progressed and as a result of biomass withdrawal, the concentration of the biomass decreased to 2 mg/L and the efficiency of purification to 50% in terms of COD. In fact, during the following days the performance of the bioreactor practically stopped, its efficiency was 15–25%. Despite the decay of the bio-system, the feeding of the bioreactor was continued and from day 380 the biomass began to grow again. On day 410 the normal performance of the bioreactor had been re-established. In comparison with the first adaptation period that lasted 120 days, the last period of adaptation was quite short – 50 days. The results of the experiments indicate that a long adaptation period makes possible developing effective activated sludge and achieving high purification efficiencies.

Figure 2 shows the effect of organic loading on the efficiency of aerobic bio-oxidation in terms of COD ( $\Delta$ COD%). Acceptably high purification efficiencies can be achieved when the loading rate applied is less than 0.2 kg/(kg day) and the hydraulic retention time is not less than 2 days. Even this efficiency does not meet the requirements of environmental protection if the concentration of contaminants in the influent is high. Decreasing the retention time or increasing the organic loading rate is not possible without deterioration in purification



**Fig. 2.** The effect of organic loading rate on the efficiency of aerobic bio-oxidation in terms of COD ( $\Delta$ COD,%).

efficiency. Thus, it is impossible to achieve sufficient reduction of contaminants in treated wastewater using only biological treatment.

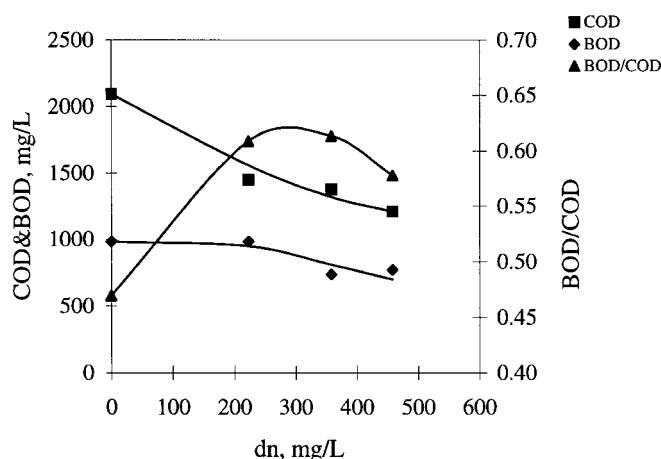
### POST-OZONATION OF BIOLOGICALLY TREATED SYNTHETIC LANDFILL LEACHATE: EXPERIMENTAL AND RESULTS

In order to improve the efficiency of purification at higher organic loadings, post-ozonation experiments were carried out using synthetic landfill leachate that had been biologically treated and had a relatively high residual COD. The effect of ozone dosage on COD and BOD removal and biodegradability of the treated leachate were studied.

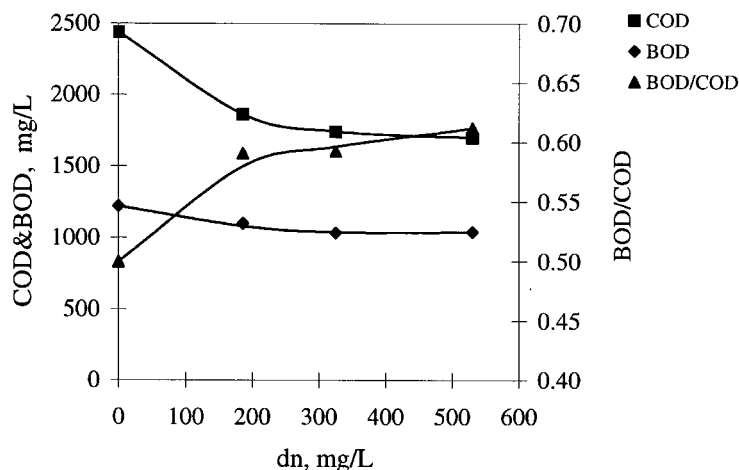
The experiments of post-ozonation were conducted in a semi-batch glass reactor (volume 0.9 L) with a continuous flow of gas through the reactor. The ozone-containing gas was added at the bottom of the reactor through a ceramic diffuser. Ozone concentrations in the influent and effluent gas streams were measured with a spectrophotometer (Specord UV-VIS). Also the gas flow rates were measured.

The experiments indicated that COD removal was 35–45% and BOD removal 30–35%. The ozone dosage needed to noticeably decrease the residual COD was 350–450 mg/L. The ratio  $\Delta\text{COD}/\text{dn}$  (removal of COD in mg/L per 1 mg/L of consumed ozone) ranged from 0.5 to 2. Biodegradability of the treated water defined as the BOD/COD ratio increased about 20% at the beginning of the ozonation and then remained essentially unchanged.

Figures 3 and 4 show the dependence of COD, BOD, and BOD/COD ratio of wastewater on the consumed ozone dosage. The wastewater was previously treated biologically at retention times of 6 and 3 days. The dependence was typical of the ozonation of all biologically treated synthetic leachates.



**Fig. 3.** The effect of ozone dosage (dn) on COD, BOD, and the ratio BOD/COD of biologically treated synthetic landfill leachate. Retention time in previous bio-oxidation 6 days.



**Fig. 4.** The effect of ozone dosage (dn) on COD, BOD, and the ratio BOD/COD of biologically treated synthetic landfill leachate. Retention time in previous bio-oxidation 3 days.

The results showed that residual organic substances can be oxidized to more biodegradable substances by post-ozonation. This makes subsequent aerobic bio-oxidation reasonable.

## CONCLUSIONS

The results of the experiments indicate that

- aerobic bio-oxidation is efficient in purifying the synthetic solution corresponding by its composition to leachates of young landfills only under stable operating conditions and if a sufficiently long retention time is applied;
- the efficiency of the post-ozonation is relatively low. However, at the beginning of post-ozonation at low ozone dosages consumed the biodegradability of the wastewater increased. It indicates the practicability of using ozone in repetitive sequential aerobic bio-oxidation and ozonation treatment of landfill leachate to achieve a required degree of purification.

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## Prügilavete puhastamine: aeroobne biooksüdatsioon ja järelsoonimine

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Puhastusprotsessi efektiivsuse parandamiseks on uuritud prügilavete puhas-  
tamist aeroobse biooksüdatsiooni ning järelsoonimisega.

Uuritavaks reoveeks oli sünteetiline prügilavesi. Reovee aeroobne bio-  
oksüdatsioon toimus pidevalt töötavas läbivoolureaktoris. Katsed näitasid, et on  
võimalik saavutada kõrget aeroobse biooksüdatsiooni efektiivsust, kui kasutada  
stabiilseid töötingimusi ning pikka hüdraulilist viibimisaega. Protsessi käigus  
vähenes biokeemilise ja keemilise hapnikutarbe suhe (BHT/KHT) kõikidel  
režiimidel. Bioloogiliselt töödeldud vee osoonimisel perioodilises kontaktsead-  
mes, s.o järelsoonimisel ilmnes, et protsessi efektiivsus on suhteliselt madal –  
jääkosooni märgatavaks vähendamiseks on vajalik suhteliselt suur osooni doos.  
Kuid juba osoonimise algetapil suurenes BHT/KHT suhe, s.t paranes reoainete  
biolagundatavus. Seega on osoonitud reovett võimalik uuesti suunata bioloogi-  
lisele töötlemisele või teha biopuhastus kombineeritult ja üheaegselt osooni-  
misega.