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Landfill leachate treatment by means of anaerobic membrane bioreactor

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Abstract

The study was undertaken to examine feasibility of biological treating of landfill leachate in anaerobic submerged membrane bioreactor. The aim of the work was to estimate the optimal concentration of leachate in the reactor influent and process parameters on the base of anaerobic digestion efficiency. The treatment efficiency under different feeding condition of leachate dilution in the range of 5–75% (v/v) with a synthetic wastewater was studied. A higher COD removal over 95% was maintained with leachate addition of 10% and 20% (v/v). Gradual decrease in organic removals was observed as leachate percentage increase. At leachate addition of 25% the COD removal reached value of 80%. For leachate concentration in influent above 30% (v/v) significant decrease of anaerobic treatment efficiency due to inhibition of microbiological activity was observed.

The influence of various hydraulic retention time (HRT) and organic loading rate (OLR) on pollutants removal efficiency and biogas production was also investigated. MBR reactor was operated at HRT in the range of 7–1 days and OLR in the range 0.7–4.9 kg COD/m^3 d. The best anaerobic digestion efficiency (COD removal 90%) was observed for HRT of 2 days and OLR of 2.5 kg COD/m^3 d.

Keywords: Anaerobic treatment; Landfill leachate; Membrane bioreactor

1. Introduction

The method of sanitary landfill for the disposal of municipal solid wastes continues to be widely used in most of European countries. The major long term problems caused by landfills are related to the generation of leachate which can cause considerable environmental problem. The composition of landfill leachate varies from site to site depending on solid waste composition, operation and hydrology of landfill, climate and the age of the landfill. In general, leachate is highly

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contaminated with organic contaminants, with ammonia, halogenated hydrocarbons, heavy metals and inorganic salts [1–3].

High loading of landfill leachate, divergent composition and different volume of leachate in particular seasons of the year make the treatment of such wastewater very complicated.

Processes for landfill leachate treatment used today are often combined techniques; usually combinations of physical, chemical and biological methods are used. Among the biological methods used for leachate treatment aerobic, anaerobic and anoxic processes are the major ones [4–7]. While air stripping, adsorption and membrane filtration belong to the major physical methods applied to leachate treatment. Among the membrane processes, reverse osmosis has been one of the most widely used methods for the last 20 years.

There is growing interest in combining membranes with biological wastewater treatment — membrane bioreactor (MBR) — where membranes are the main solid–liquid devices [8]. There are two types of MBR reactors according to the locations of membrane units i.e. submerged and external reactors. In the recent years submerged membrane reactors have attracted great attention due to more compact system and energy saving. The submerged membrane bioreactor is an improvement on the conventional activated sludge process, where the traditional secondary clarifier is replaced by membrane unit of treated wastewater from the mixed solution in the bioreactor [9].

Membrane bioreactor process has been one of the alternatives for both municipal and industrial wastewater treatment. MBRs use ultrafiltration and/or microfiltration membranes for the completion retention of sludge. This leads to increase microorganism's concentration in the reactor and improvement process efficiency with lower sludge production. The industrial application of membrane bioreactors include i.e. oil wastewater treatment, nitrogen removal from food processing wastewater and complex compounds from pharmaceutical wastewaters.

In this study, anaerobic submerged membrane bioreactor was employed for landfill leachate treatment. The aim of the work was to estimate the optimal concentration of leachate in the reactor influent on the base of anaerobic digestion efficiency. The influence of various hydraulic retention times and organic loading rate on pollutants removal efficiency and biogas production was also investigated. Due to poor quality of MBR effluent the RO process and stripping has been employed to post-treatment.

2. Materials and methods

2.1. Membrane bioreactor

The 29 L capacity laboratory scale submerged membrane reactor with the capillary ultrafiltration module (Zenon) was used in this study (Fig. 1). A nominal pore size of membranes was 0.1 μm and effective filtration area was 0.46 m^2.

The reactor was filled up with granular sludge from industrial wastewater treatment plant and increasing percentage of leachate. Biological treatment of leachate under anaerobic conditions was conducted using anaerobic granular sludge at the concentration of 10 g/L. Experiments were performed under temperature of 35°C.

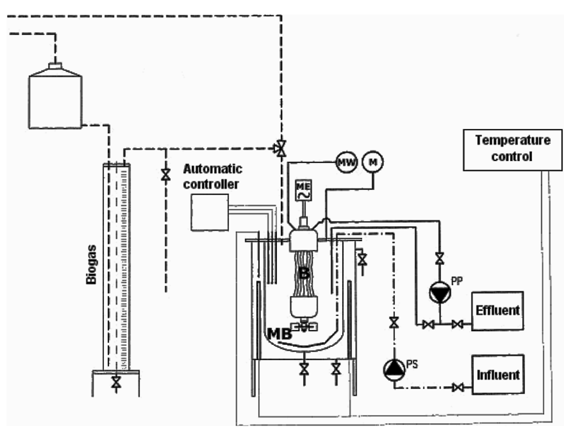

Fig. 1. Schematic diagram of experimental system (MB-membrane bioreactor, B-membrane module).

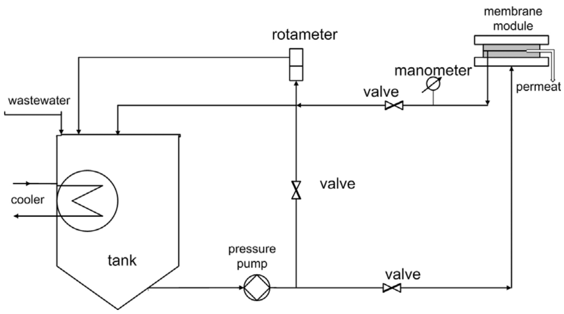

Fig. 2. Laboratory scale membrane test unit.

2.2. Reverse osmosis unit

The effluent from anaerobic bioreactor was post-treatment using reverse osmosis. The RO process was carried out on laboratory scale membrane test unit illustrated in Fig. 2. The specification of the RO membranes is given in Table 1.

2.3. Analytical methods and leachates

Standards methods [10] were used for measurement of total solids (TS), volatile solids (VS), volatile fatty acids (VFA), ammonia nitrogen (NH_4^+), chloride, total alkalinity and pH. Chemical oxidation demand was analyzed colorimetrically using tests and photometer of the HACH firm (DR 4000). The percentage of methane and carbon dioxide were analyzed by a gas chromatograph (Shimadzu). Chemical composition of synthetic wastewater was prepared according to PN-72/C-04550. It was also calculated a methane specific yield according to the following formula:

$$Y = \frac{V_b}{a}[\mathrm{dm^3 g^{-1} COD_{removed}}] \quad (1)$$

where
V_b, biogas production per day (dm^3/d)
a, COD removal per day (g/d).

The specific values of COD for influent and bioreactor effluent were measured at 2- to 4-day intervals. Redox potential, pH and collected biogas were monitored daily.

The leachate employed was obtained from the Sobuczyna municipal landfill located close to the urban area of the city Czestochowa in the southern Poland. The Sobuczyna landfill was started in 1987 and was still in use during the time of this study. The landfill is a traditional landfill receiving both household wastes and industrial wastes. This is an old landfill and leachates from this tip present relatively low COD values and low ratio of BOD/COD compared with COD values from young landfills.

Table 1
Specification of RO unit

Parameter	Membrane "Osmonics" Inc. type SEPA CF — HP, RO-DS3SE
Pressure, MPA	Max 4.0
pH, range	1–11
Temperature, °C (max)	90
Sodium rejection, (%)	C = 0.2% NaCl R_{NaCl} = 98.9
Membrane material	Polyamide

3. Results and discussion

3.1. Landfill leachate characterization

Table 2 shows the average values of the composition of these leachates during experimental period. COD values were low, due to the age of landfill. Ammonia nitrogen content was high and was directly related to basic pH.

Table 2
Composition of Sobuczyna landfill leachate

Parameter	Unit	Min–max
COD	mg/dm^3	2800–5000
pH	–	8.0–8.9
Alkalinity	mg/dm^3	4600–7900
Chloride	$mg/dm^3\ Cl^-$	1950–3650
Ammonia	$mg/dm^3\ NH_4^+$	750.4–840.0

3.2. Anaerobic leachate treatment

In the first step of the experiment the membrane bioreactor was seeded with anaerobic granules and fed with high loaded synthetic wastewater in order to allow bacterial community to acclimatize. MBR reactor was operated at organic loading rate (OLR) of 1.0 kg COD/m^3 d, and hydraulic retention time (HRT) of 7 days. After four weeks acclimatization process the COD removal efficiency achieved value of 95% (Fig. 3). Concentration of organic compounds in effluent measured as chemical oxygen demands has been stabilized on the level of 385 mgO_2/dm^3 while daily biogas production achieved value of 12 L.

After acclimatization process the maximum percentage of leachate that can be biologically treated without inhibition of microbiological activity. The digestion efficiency under different feeding condition of leachate dilution in the range of 5–75% (v/v) with a synthetic wastewater was studied. The main results are shown in Fig. 4.

Gradual decrease in organic removals was observed as leachate percentage increase in MBR influent. A higher COD removal over 90% was

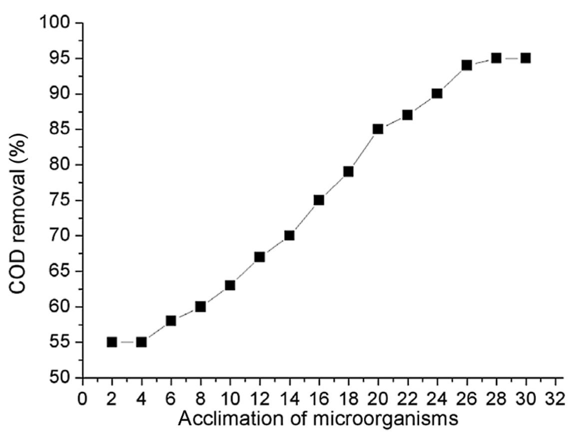

Fig. 3. Variations of COD in MBR effluent during adaptation process.

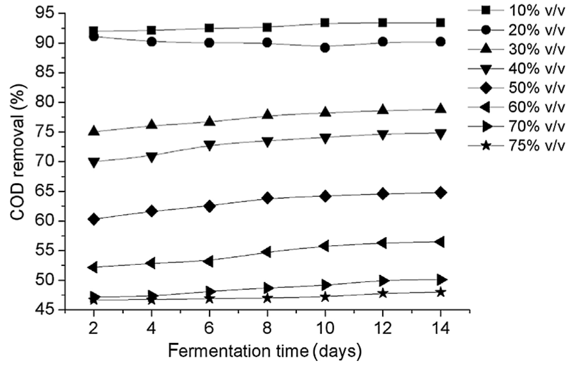

Fig. 4. Relationship between COD removal and leachate concentration in MBR influent.

maintained with leachate addition of 10% and 20% (v/v). For leachate addition of 30% (v/v) anaerobic treatment efficiency dropped to the level of 78.8%. At leachate addition higher then 30% organic removal gradual decreased and achieved value of 45% for leachate concentration of 75% (v/v).

Gradual decrease in biogas production was observed as leachate percentage increase. Specific methane yield calculated for conducted experiments varied in the range of 0.45–0.35. The effluent redox potential changed from −466 mV to −417 mV. There was no significant variation of pH (7.8–8.4) and alkalinity during fermentation process. The membrane effluent was free of suspended solids. During all experiments the VFA/alkalinity ratio, which shows fermentation properly was estimated. It is assumed the maximum value above which process inhibition is observed is on the level of 0.3. The highest value of VFA/alkalinity ratio (0.27) was observed in the experiment with leachate content in influent of 75% (v/v) while for others fermentation processes the ratio achieved constant level, in the range of 0.16–0.25.

The leachate percentage content of 20% (v/v) was selected as the most appropriate in MBR influent and was used for the rest of the study.

4. Effects of organic loading and hydraulic retention time on the MBR system

HRT was a very important parameter in the submerged membrane bioreactor due to influence on treatment efficiency as well as size of bioreactor and engineering design [11,12]. In this study the MBR system was operated with mixture of leachate and synthetic wastewater under HRTs of 7, 5, 3, 2, 1.5 with increase of OLR from 0.7 kg/m^3 × d to 4.9 kg/m^3 × d respectively. The COD removal efficiency under HRT of 7 days and OLR of 0.7 kg/m^3 × d achieved value of 76%. For the shorter hydraulic retention time (5 d) the treatment efficiency was 2% higher and effluent COD value was around 1089 mgO$_2$/dm^3. Moreover a daily biogas production was 36% higher than that under the highest HRT of 7 d. There was no significant variation of redox potential and pH in the reactor

During fermentation process under HRT of 3 days gradual increase of treatment efficiency was still observed. Effluent COD reduced to the level of 940 mgO$_2$/dm^3 and a daily biogas production was two times higher than that under the HRT of 3 d (13,470 cm^3). Increase of biogas yield to value of approximately 0.35 dm^{3}biogas/gCOD$_{remover}$ × d and redox potential in MBR reactor was also observed.

Shortening HRT to 2 days caused increase of OLR to 2.5 kg/m^3 × d. Under such conditions treatment efficiency was *n* the level of 90% and effluent COD decrease to value of 417 mgO$_2$/dm^3. Moreover a daily biogas production was 2 times higher than that under the HRT of 2 d. Gradual decrease of redox potential in membrane reactor was still observed (370 mV).

At HRT of 1.5 d and OLR of 3.3 kg/m^3 × d the organic removal efficiency was 13% lower as compared to treatment process under HRT of 2 d. It was found worse effluent quality (COD$_{effluent}$ 1380 mgO$_2$/dm^3) and lower biogas production. The concentration of volatile fatty acids in the reactor was 50% higher than in anaerobic process

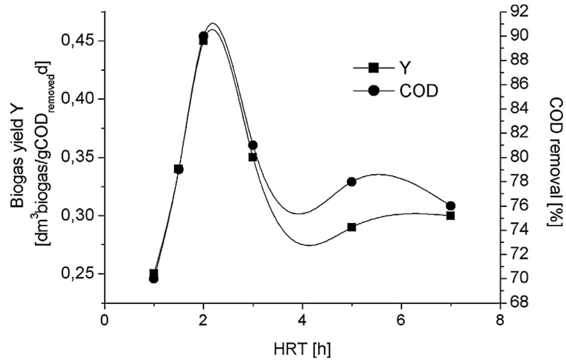

Fig. 5. Relationship between COD removal efficiency, daily biogas production and hydraulic retention time.

under HRT of 2 d. Due to increase of redox potential in the reactor up to −340 mV a decrease of daily biogas production was observed. Biogas production during fermentation process is determined by proper redox potential in the range of −350 to −450 mV; for the higher redox value i.e. −250 to −300 mV metanogenesis is completely inhibited.

Shortening HRT to 1 day and increase of organic loading rate to 4.9 kg/m^3 × d worsened the organic removal efficiency to the level of 30% lower as compare to the process under HRT of 1.5 d. The concentration of volatile fatty acids

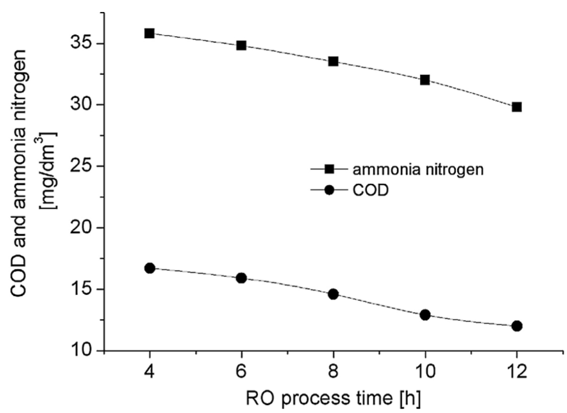

Fig. 6. COD and ammonia removal efficiency during RO post-treatment process.

Table 3
Efficiency of landfill leachate treatment in MBR and RO processes

Parameter	Unit	Raw wastewater	Permeate from MBR reactor	Permeate from RO unit	Permisaible standards[a]
COD	mg/dm^3	5000	417	12	125
pH	–	8.03	8.18	8.9	6.5–9
Ammonia	mg/dm^3	381.5	206	29.8	10

[a]Regulation of the Ministry of Environmental Protection, Natural Resources and Forestry, dated 8 July 2003, on the classification of water and conditions the sewage discharged to waters and soil should satisfy, Journal of Law No.168, item 1763.

achieved value of 1560 $mg/dm^3 CH_3COOH$, that is close critical level for fermentation process. Also critical value for redox potential was observed in the bioreactor (−320 mV).

Very important parameter monitored during cotreatment of leachate and synthetic wastewater was biogas yield. At the longer HRT of 7 d and 5 d, the biogas yield was on the same level (0.3 dm^3biogas/$gCOD_{remover} \times$ d) whereas the highest value achieved at HRT of 2 d (0.45 dm^3biogas/$gCOD_{remover} \times$ d). Relationship between COD removal efficiency, daily biogas production and hydraulic retention time was presented on Fig. 5.

4.1. Post-treatment in RO process

Because of poor quality of effluent from MBR reactor (COD-417 mgO_2/dm^3, ammonia - 206 mg/dm^3 NH_4^+), the reverse osmosis has been employed as a post-treatment technique. In order to determine transport properties of used membranes they have been tested with distillated water determining a relationship between the volume water flux and transmembrane pressure from 1.0 MPa to 3.0 MPa. It was found that volume water flux increased with increasing pressure. Permeate flux achieved value of 8.960×10^{-6} m^3/m^2 s at transmembrane pressure of 2 MPa and cross flow velocity of 2 m/s. Under such conditions the post-treatment process has been carried out for the MBR effluent and results of that experiment are presented on Fig. 6.

Comparison of treatment efficiency of anaerobic co-treatment in MBR reactor and RO post-treatment process were shown in Table 3.

Although the treatment efficiency in RO process was very high permeate cannot be discharged into natural water because of high concentration of ammonia (29.8 mg/dm^3). That is why in last step of this study the stripping process has been applied. Permeate from RO has been aerated for 12 h at pH 12. The efficiency of striping process achieved value of 76% with effluent ammonia concentration of 7.1 mg $N\text{-}NH_4^+/dm^3$.

5. Conclusion

In this study biological treating of landfill leachate in anaerobic submerged membrane bioreactor was examined. The most important results are:

- It was possible to acclimatize a bacterial population for the treatment of landfill leachate in laboratory scale MBR reactor .The leachate percentage content of 20% (v/v) is the most appropriate in MBR influent.
- The most favorable COD removal of leachate and synthetic wastewater mixture was 90% at organic loadings of 2.5 kg/m^3 d and at an HRT of 2 d.

- Due to poor quality, permeate from MBR reactor can not to be discharged into natural water without additional treatment process. RO and stripping processes are suitable for post-treatment of membrane bioreactor effluent.

Acknowledgement

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