Study on Anaerobic Landfill Leachate Treatability by Membrane Bioreactor

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Abstract— This study was undertaken to examine feasibility of biological treating of landfill leachate that was collected from Tehran municipal landfill with about 0.5 – 1 year old. Average COD of the leachate in aerobic submerged membrane bioreactor is 68000 mg/L. The reactor with a working volume of 175 L, having membrane module (Hollow fiber) with pore size of 0.1 µm was used in this study. The dissolved oxygen concentration (DO) was maintained at 3.2 mg/L and solid retention times (SRTs) and hydraulic retention times (HRT) were controlled at 55 and 15 days respectively. Following that the mixed liquor suspended solids (MLSS) was stabilized at 6.3 g/L. Under condition of COD/N ratio of 46 and COD/P03-P ratio of 455 and BOD/COD ratio of 0.65, the average effluent COD was 1733 mg/L with average removal efficiency of 97.46%. The average NH4 –N removal efficiencies was 99% which was reached to 99.7% at maximum level.

Keywords—Municipal landfill; leachate; Membrane bioreactor; Nutrient removal

I. INTRODUCTION

The method of anaerobic sanitary landfill for the disposal of municipal solid wastes continues to be widely used in the most countries in the world [3, 17 and 19]. Leachate is a high-strength wastewater formed as a result of percolation of rain-water and moisture through waste in landfills [1]. During the formation of leachate, organic and inorganic compounds are transferred from waste to the liquid medium [21] and pose a hazard to the receiving water bodies. Production of landfill leachate begins with introducing moisture waste into disposal area and continues for several decades following the landfill closure. Leachate contains high organic matter and ammonium nitrogen and varies from site to site and its composition depends upon the landfill age, the quality and quantity of waste, biological and chemical processes that took place during disposal, rainfall density, and water percolation rate through the waste in the landfill. Depending upon what was placed in the landfill, leachate may contain many types of contaminants, and if not removed by treatment, these contaminants may be toxic to life or simply alter the ecology of receiving streams. Leachate should be treated before reaching surface water or ground water bodies, because it can accelerate algae growth due to its high nutrient content, deplete dissolved oxygen in the streams, and cause toxic effects in the surrounding water life. Since the composition of a leachate consists of a wide range of contaminants, it cannot be easily treated by conventional methods. Therefore, a number of scientists around the world have intensively focused on the combination of biological and physico-chemical treatment systems for effective leachate treatment [18, 1].

Alternative treatments have been reviewed. Briefly, leachate can be recirculated to the same landfill or treated by different methods: biological, aerobic, anaerobic methods and/or nitrification–denitrification to remove organic matter and ammonium nitrogen. Biological processes to remove organic matter can be effective for young leachate with a high BOD5/COD ratio [2]. Many researchers reported that the membrane bioreactors are effective treatment alternatives for the young leachates [1] Landfill leachate is characterized by its generation rate and composition, both of which are affected by the age of the landfill site [22]. In particular, Leachate consists of many different organic and inorganic compounds that may be either dissolved or suspended and which are biodegradable and non-biodegradable [22]. In addition to this, the characteristic of the leachate varies with regard to its composition and volume, and biodegradable matter present in the leachate with time. For this reason, young and old landfill leachates have very different features. Calace et al. reported that the young landfill leachate fractions have low molecular weight distributions (<500 Da) at the rate of 70%, while the high molecular weight distribution (>10,000 Da) is 18%. Besides, the low and high molecular weight distributions are 28 and 67%, respectively, in old landfill leachate samples [3]. According to this result, easily biodegradable components of leachate reduce, and constituents having high molecular weights and that are nonbiodegradable increase in the course of time. These factors make leachate treatment difficult and these factors needed to be taken into account when different treatment processes are considered. The treatment requirements for leachate from sanitary landfills can vary depending on the discharge limits and contaminants present [3].

Among advanced biological treatment processes, membrane bioreactor (MBR) is the most important process [1]. MBR are considered as a good integration of conventional activated sludge (CAS) system and advanced membrane separation, thus enabling the independent control of sludge retention time (SRT) and hydraulic retention time (HRT) and retaining a high concentration of sludge biomass.
in the reactors. Compared with CAS processes, MBR process has great advantages including a smaller footprint, less sludge production and better effluent quality [23].

MBR can be operated at very long sludge ages and can extend greatly the field of application of biological processes for concentrated streams, such as leachate [18]. The combination of membrane separation technology and bioreactors has led to a new focus on wastewater treatment. It contributes to very compact systems working with a high biomass concentration and achieving a low sludge production with an excellent effluent quality. Membrane bioreactors have been widely applied at full scale on industrial wastewater treatment and some plants have been adapted to leachate treatment [7, 8]. The process efficiencies were in the range of 95–98% in terms of TOC reduction, and exceeded 97% for specific organic pollutants. Contrary to conventional systems, organisms such as nitrifiers or organisms which are able to degrade slowly biodegradable substances are not washed out of the system and no loss of process activity occurs [8].

II. MATERIALS AND METHODS

A. leachate feed

The leachate used in this study was collected from a municipal landfill located in Kahrizak near Tehran city in the Tehran province. The landfill has been in operation since 1985, the old of landfill for sampling is 0.5-1 years old. The large volume samples (200 L) were collected and stored in a retention tank every week. The characteristics of the landfill leachate investigated are shown in Table 1. Leachate used during this study was young because it contained readily biodegradable organic matter [1].

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>COD, mg/L</td>
<td>68230±8000</td>
</tr>
<tr>
<td>BOD, mg/L</td>
<td>4450±3600</td>
</tr>
<tr>
<td>NH3-N, mg/L</td>
<td>1470±90</td>
</tr>
<tr>
<td>NO3-N, mg/L</td>
<td>150±50</td>
</tr>
<tr>
<td>pH</td>
<td>6.9±0.2</td>
</tr>
<tr>
<td>PO4-P, mg/L</td>
<td>130±40</td>
</tr>
<tr>
<td>BOD/COD</td>
<td>0.65</td>
</tr>
<tr>
<td>COD/N</td>
<td>46.42</td>
</tr>
<tr>
<td>CI-, mg/L</td>
<td>14800±1000</td>
</tr>
<tr>
<td>SO4, mg/L</td>
<td>5500±300</td>
</tr>
<tr>
<td>Conductivity, mS/m</td>
<td>44±6.5</td>
</tr>
<tr>
<td>Turbidity, Ntu</td>
<td>3896±270</td>
</tr>
</tbody>
</table>

B. process configuration and system design

The investigations were carried out at a laboratory scale in a MBR reactor. The reactor with a working volume of 175 L was made of Plexiglas. Dissolved oxygen was supplied using 2 fine bubble disc diffusers (Ecoflex 250 CV). Made by USA diffuser tech Co. Placed at the bottom of the reactor, producing bubbles of pour size. The amount of oxygen supplied to the reactor was regulated in order to maintain the oxygen concentration at a level of 3 mg/L, one blower pump with capacity of 190 m3h-1 and pressure of 320 mbar, supplied system air requirement, an adjustable air valve controlled.

To startup the reactor, we used the returned activated sludge of Gitarieh wastewater treatment plant, which is located in north of Tehran. After each time of aeration rates changes we gave minimum 55 day (SRT) to system to adapt with new condition, analyses were done, when the system reached steady state condition, MLSS at the end of operation in the reactor is assumed as the steady state index, samples were taken from feed and treated leachate, samples were stored in polyethylene cap bottles with capacity of about 100 ml, temperature and dissolved oxygen was measured online air volumetric flow rate in aeration reactor was measured by a volumetric contour in a defined time.

Due to importance of timely stages control in MBR system as well as setting feeding, vacuuming, backwashing and measuring and recording of dissolve oxygen and temperature, we used a PLC and computerized system with essential accessories, includes control and relays boards, dissolved oxygen and temperature probes, electrical valves, feed pump, figure 1 present system schematic, which had a working volume of approximately 175 L, in which the membrane module was directly submerged, is shown in Fig. 1. The dissolved oxygen concentration (DO) was maintained at 3.2 mg/L by adjusting the air flow to 4 m³/h/L. The water level in the bioreactor was controlled with a level controller and a level sensor. The concentration of the mixed liquor suspended solids (MLSS) was 6300 mg/L. The sludge was withdrawn continuously with a pump set at different solid retention times (SRTs). HRT was controlled at 15 days by a rotary flow meter under the operational condition of invariable membrane flux, The effluent of the bioreactor was connected to an automatic vacuum effluent system directly by a rotary flow meter. When the vacuum pump started, part of the gas in the MBR was pumped out to create a negative pressure, and then the wastewater in the bioreactor was drained out through the membrane module and entered into the treated wastewater tank. Backwash electrical gate valve (1 minute in 1 hour) and vacuum pump (3 minutes in 1 hour) periodically run in the automatic vacuum effluent system. However minimum vacuum level was kept by a floater, whether feeding or vacuum pumps were operating or not. Therefore, wastewater noncontinuous entered the bioreactor through the flowed into the submerged membrane module. Feeding pump operate noncontinuous at operating time.

C. Membrane characteristics

In this pilot-plant test, a hollow-fiber pp microfiltration (MF) membrane was used with pore size of 0.1µm and the effective surface area of a MF membrane module at 4m². Membrane flux was between 0.5 and 0.8 m³/d.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Hollow fiber</th>
</tr>
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<tbody>
<tr>
<td>Raw material</td>
<td>PP</td>
</tr>
<tr>
<td>Diameter inside</td>
<td>320-350µm</td>
</tr>
<tr>
<td>Diameter outside</td>
<td>400-450µm</td>
</tr>
<tr>
<td>Pore size</td>
<td>0.1-0.2µm</td>
</tr>
<tr>
<td>Pore density</td>
<td>40-50%</td>
</tr>
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</table>
A lab-scale MBR was set up for COD and biological nutrient removal with a flow rate of 12 L/day, and operated under a total SRT of 55 days, and an HRT of 15 days. Influent characteristics were measured once per week. The effluent BOD5, COD, NO3-N, and PO4-P, NH4-N, NO2-N, EC, turbidity, Cl-, SO4 were monitored for six weeks. All experiments were conducted under conditions of constant temperature (21 ºC) and controlled pH (9.5). Determination of COD, BOD5, nitrite, nitrate, ammonium, phosphate, sulfate, CL-, conductivity, MLSS and turbidity were performed according to standard methods, 20th ed. [4]. Were used for measurement of total solids (TS), volatile solids (VS), volatile fatty acids (VFA), ammonia nitrogen (NH4), chloride, total alkalinity and pH. Chemical oxidation demand experiments were conducted under conditions of constant effluent BOD5, COD, NO3-N, and PO4-P, NH4-N, NO2-N, temperature (21 ºC) and controlled pH (9.5). Determination of COD, BOD5, nitrite, nitrate, ammonium, phosphate, sulfate, CL-, conductivity, MLSS and turbidity were performed according to standard methods, 20th ed. [4] Were used for measurement of total solids (TS), volatile solids (VS), volatile fatty acids (VFA), ammonia nitrogen (NH4), chloride, total alkalinity and pH. Chemical oxidation demand was analyzed colorimetrically using tests and photometer of the HACH firm (DR 2010).

### III. RESULTS AND DISCUSSION

#### A. COD removal

The average COD concentrations in the influent are 68000 mg/L. During the operation, removal efficiencies of COD were above 97.0% suggesting that it was irrespective of COD/N ratios. With high COD/N ratio of 45, the nutrient requirements decrease as the sludge age increases because net sludge production decreases as sludge age increases generally, for sludge ages greater than 10 days, the nitrogen removal attributable to net sludge production is less than 40 mg COD/mg N applied [5]. The maximum value for percent COD removal was obtained around COD/P-PO4 =160 and then dropped above 160 indicating phosphate limitation at high COD [10]. Increasing COD/N-NH4 ratios from 10 to 50 resulted in decreases in COD removal efficiencies steadily possibly because of ammonia and PO4 limitations at high COD/N-NH4 and COD/P-PO4 ratio [10]. In this study the sludge age (SRT) regulate at 55 days then the MLSS was stabilized at 6.3 g/L. Despite the fluctuations of average influent COD concentration ranging from 60000 mg/L to 75000 mg/L, the effluent COD concentrations were always lower than 1935 mg/L, as shown in Fig. 2 (a). The average effluent COD concentrations was 1733 mg/L, with the average efficiency of 97.46%, under COD/N ratio of 46 and COD/P-PO4 ratio of 455 and BOD/COD ratio of 0.65 These data indicated that the system could provide a consistent high efficiency of COD removal.

#### B. Nitrogen removal

Removal efficiency for ammonium-nitrogen increased with COD/N-NH4-N ratio between 10 and 40, because of high ammonium concentrations at low COD/N-NH4-N ratio, ammonium removal efficiencies were low [10]. Increasing COD/P-PO4-P ratio resulted in steady increases in NH4-N removal efficiencies for COD/P-PO4-P values between 40 and 250 indicating adverse effects of high phosphate levels or COD limitations at low COD/P-PO4-P values [10].

During the whole operation, the removal efficiencies of NH4–N and TN were as good as COD removal, as shown in Fig. 2 (b). During the operation of COD/N is 46.42, the NH4–N and TN concentrations removal in effluent were almost the same, indicating no NOx–N accumulation was observed in the reactor. Produces of nitrification were almost completely denitrified to nitrogen gas. The average NH4–N and TN removal efficiencies were 99% while the average NH4–N effluent concentrations were 10 mg/L, initially, nitrification activity increased gradually, and the highest NH4–N removal efficiency was 99.7%. However, an almost completely nitrification was achieved during this period. These data showed that 40.55 mg/L NOx–N was removed after 30-40 days, which implying that the denitrification was accomplished completely.

From 20 d to 40 d, the effluent NH4–N concentration was increased to above 10 mg/L. Initially, the effluent NH4–N and TN concentrations increased drastically, due to that the microorganism need a period to acclimate the change. Afterwards, effluent concentrations were decreased gradually, suggesting that the nitrification and denitrification capacities were strengthened gradually.

#### C. PO4-P removal

Increasing COD/P-PO4-P values from 40 and 160 resulted in increases in phosphate removal efficiency. Because of excess phosphate or COD limitations at low COD/P-PO4-P values the phosphate removal efficiency was low. The efficiency decreased for COD/P-PO4-P values above 200 because of phosphate limitations at high COD/P-PO4-P values [10].

Fig. 2 (c) presents the PO4–P concentrations in the effluent during the whole operation period. With COD/P-PO4-P ratio of 455, although the PO4–P concentration in the effluent was low and the average PO4–P removal efficiency was as high as 90%, with increasing COD/N ratio, the PO4–P concentrations in anoxic and aerobic zones decreased insignificantly, and effluent concentration between 1d and 20d was as high as 23.25 mg/L and the average PO4–P removal efficiency was 85.3%, especially during operation period The PO4–P removal process was stabilized between 20 d and 40 d, with average PO4–P effluent concentration of 6.1 mg/L and removal efficiency of 96%. Additionally, PO4–P release process ceased in anoxic zone. Results indicated that low COD/N ratio decreased PO4–P removal efficiency. And increasing sludge age increasing the PO4_3–P removal efficiency.

### IV. CONCLUSIONS

The present study contains results of landfill leachate treatability by membrane bioreactor that is important for modeling, design, and operation of landfill leachate treatment MBRs and determination of discharge limits.

Suitable treatment strategy depends on two major criteria: 1. the initial leachate quality and 2. the final requirements given by local discharge water standards [17] the presented data indicate that the landfill leachate composition has a

<table>
<thead>
<tr>
<th>Membrane area</th>
<th>4 m²</th>
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<tbody>
<tr>
<td>Pressure minus</td>
<td>0.01-0.03 Mpa</td>
</tr>
<tr>
<td>pH</td>
<td>0–14</td>
</tr>
<tr>
<td>Available temp</td>
<td>4–45 ºC</td>
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</tbody>
</table>
significant effect on treatability, in the young landfills and composting units leachate that organics concentration (expressed COD), BOD/COD, Ph, COD/NH4-N ratio is high. The membrane bioreactor has to operate with sludge age greater than 50 days, hydraulic retention times (HRT) 15 days. MBR was developed and demonstrated herein to treat landfill leachate. In order to evaluate the biological treatability of the landfill leachate, the removal efficiency of COD, NH4-N and PO4 –P were investigated. These data indicated that The system provided high removals in terms COD, NH4-N and PO4 –P were 1733, 10 and 13 mg/L, respectively, all of them over the permissible limit for IRAN discharge standards (COD < 200 mg/L, NH4-N < 2.5 mg/L, PO4 –P< 6 mg/L), quality of the effluent from MBR system was not appropriate (COD < 200 mg/L, NH4-N < 2.5 mg/L, PO4 –P< 6 mg/L), however after treatment in the MBR.

ACKNOWLEDGMENT
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REFERENCES
Figure 1. Schematic diagram for MBR process

Figure 2. COD concentrations in the effluent and removal in MBR (a) NH4-N concentrations in the effluent and removal in MBR (b) PO4-P concentrations in the effluent and removal in MBR (c)