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### LANDFILL LEACHATE TREATMENT USING SEQUENTIAL ANOXIC MOVING BED BIOREACTOR/AEROBIC CONTINUOUS STIRRED TANK REACTOR

### ARDIŞIK ANOKSİK HAREKETLİ YATAK BİYOREAKTÖR / AEROBİK SÜREKLİ KARIŞIMLI TANK REAKTÖR KULLANILARAK ÇÖP SIZINTI SULARININ ARTIMI

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#### ABSTRACT

This study was aimed to investigate the performance of sequential anoxic moving bed bioreactor (AnoxMBBR) and aerobic continuous stirred tank reactor (AeCSTR) to remove chemical oxygen demand (COD) and ammonium-nitrogen from landfill leachate. The sequential AnoxMBBR /AeCSTR system was tested at constant hydraulic retention time (HRT) of 48. The performance of this system was evaluated in terms of chemical oxygen demand (COD), ammonium (NH<sub>4</sub><sup>+</sup>), nitrite (NO<sub>2</sub><sup>-</sup>) and nitrate (NO<sub>3</sub><sup>-</sup>) parameters. In AnoxMBBR, nitrate removal (40%) was limited due to low COD removal efficiency. In AeCSTR, COD and ammonium removal efficiencies were obtained as 46% and 71%. Also, partial nitrification in AeCSTR was simultaneously achieved with nitrite accumulation of 1660 mg/L. The results indicated that the AnoxMBBR/AeCSTR system is quite efficient to remove COD and ammonium from landfill leachate, however, the effluent COD and ammonium concentration still did not meet the discharge criteria.

**Keywords:** continuous stirred tank reactors, denitrification, landfill leachate, nitrification, moving bed bioreactor

#### ÖZET

Bu çalışmada, çöp sızıntı sularından kimyasal oksijen ihtiyacını(KOİ) ve amonyum-azotu gidermek için ardışık anoksik hareketli yatak biyoreaktörün (AnoxHYBR) ve aerobik sürekli karışimli tank reaktörün (AeSKTR) performansını araştırmak amaçlandı. Ardışık AnoxMBBR/AeCSTR sistemi, 48'lik sabit hidrolik bekleme süresinde (HRT) test edildi. Bu sistemin performansı, kimyasal oksijen ihtiyacı (KOİ), amonyum (NH<sub>4</sub><sup>+</sup>), nitrit (NO<sub>2</sub><sup>-</sup>) ve nitrat (NO<sub>3</sub><sup>-</sup>) parametreleri açısından değerlendirildi. AnoxHYBR'de, düşük KOİ giderim verimi nedeniyle nitrat giderimi (% 40) sınırlıydı. AeSKTR'de KOİ ve amonyum giderim verimleri sırasıyla % 46 ve % 71 olarak elde edildi. Ayrıca, AeSKTR'de 1660 mg / L nitrit birikimi ile kısmi nitrifikasyon sağlandı. Sonuçlar, AnoxHYBR / AeSKTR sisteminin çöp sızıntı sularından KOİ ve amonyum giderimi için oldukça verimli olduğunu, ancak çıkış KOİ ve amonyum konsantrasyonunun hala deşarj kriterlerini karşılamadığını göstermiştir.

**Anahtar Kelimeler:** çöp sızıntı suyu, denitrifikasyon, nitrifikasyon, sürekli karışimli tank reaktör, hareketli yatak bioreaktör

## INTRODUCTION

Landfill leachate is usually characterized by high chemical oxygen demand (COD), high ammonia-nitrogen, biological oxygen demand (BOD), BOD<sub>5</sub>/COD ratio, heavy metals and strong color (Renou et al., 2008; Luo et al., 2018). Owing to these characterizations, landfill leachate (LFL) are a quite complex wastewater that causes adverse and dangerous effects on aquatic life, soil, sub-soil, groundwater and surface water (Eggen et al., 2010). This wastewater should be treated before directly released into environment, due to the fact that it includes especially a large amount of ammonium and organic matter, which causes eutrophication and dissolved oxygen consumption in natural water (Atmaca, 2009; Wang et al., 2010). Therefore, many researchers used physico-chemical methods and biological methods to remove ammonium-nitrogen and organic matter from LFL (Gkotsis, 2018; Mohajeri et al., 2019). However, the biological treatment is the most cost effective alternative over physico-chemical methods due to its less sludge production and high ammonium/organic matter removal efficiency. Although aerobic, anoxic and anaerobic biological treatment processes are commonly used, these processes alone are insufficient to remove ammonium nitrogen and organic matter from LFL. Therefore, sequential Anoxic/Oxic (A/O) process has been proposed recently and this process was effectively used to treat of the ammonium-nitrogen and organic matter rich wastewater (Zhang et al., 2015; Liu et al., 2018). Especially, this sequential process offers advantages such as saving of added organic carbon for anoxic denitrification and less oxygen consumption for aerobic nitrification. With high COD concentration of leachate, however, microbial population in Anoxic stage of sequential process causes usually inhibition.

The various reactor configurations can be effective on shock loading during operation of A/O systems. Hence, many researchers have been focused on LFL treatment performance of attachment and suspended biological reactor configurations (Chen et al., 2018; Zhang et al., 2018; Xiong et al., 2018; Liu et al., 2017). The CSTR are feasible and eco-friendly technologies in organic carbon and ammonium nitrogen removal. The use of CSTR process to treat LFL can contribute to removal of organic matter and ammonium nitrogen (Ağdağ et al., 2005). Additionally, the MBBR process by using carriers in which microorganism forms biofilm is preferred over other biological reactor configurations due to advantages such as simple construction, low space requirement, low sludge production, high biomass concentration and long sludge residence time (Kawan et al., 2016). In recent years, few researchers have reported that MBBR process is effectively used for nitrification and denitrification of municipal sewage (Malovanyy et al., 2015). However, there are limited studies on LFL treatment using sequential AnoxMBBR-AeCSTR process in literature.

In this context, the main aim of this study was to investigate the effectiveness of sequential anoxic MBBR–aerobic CSTR systems to remove simultaneous organic matter and nitrogen from real LFL.

## MATERIAL AND METHODS

### *Landfill Leachate and Inoculated Sludge*

The both bioreactors had already been operated in batch-mode under anoxic and aerobic conditions for more than 80 days. Therefore, the microbial population had already been well acclimated to the heterotrophic denitrification and the aerobic nitrification conditions. Additionally, the mixed liquor suspended solids (MLSS) concentrations of AnoxMBBR and AeCSTR were around 6 g/l and 10 g/L, respectively.

The LFL was monthly taken from landfill site in Kahramanmaraş, Turkey. The characteristics of LFL used throughout the study are illustrated in Table 1.

**Table 1.** Characteristics of the LFL

Parameter	Concentration*
pH	7.98±0.1
COD	10550±350 mg/L
NH <sub>4</sub> <sup>+</sup>	1620±75 mg/L
NO <sub>3</sub> <sup>-</sup>	55±5 mg/L
NO <sub>2</sub> <sup>-</sup>	1±0.1 mg/L

\*Values are average of triplicate measurements

### ***Set-up and Operation of Reactors***

In this study, the two glass bioreactors (Bioflo 110, New Brunswick Scientific Co, Edison, NJ, USA) with an activate working volume of 5 L were operated as AnoxMBBR and AeCSTR. In anoxic reactor, AnoxKaldnes K1 carrier material was used as biomass carrier with volumetric filling ratio of 40%. The AnoxMBBR was fed with raw LFL and AeCSTR was fed with AnoxMBBR effluent. The influent of the AnoxMBBR was supplemented with about 650 mg/L  $\text{NO}_3^-$  as the external electron acceptor source of the heterotrophic denitrification process, while organic carbon in LFL was used as electron donor source. The both reactors were mixed via a shaft impeller system. The temperature of AnoxMBBR and AeCSTR were kept at  $30\pm 1^\circ\text{C}$  and  $25\pm 2^\circ\text{C}$ , respectively. In AeCSTR, dissolved oxygen (DO) concentration over 4 mg/L was provided via an air pump (Resun Air Pump LP-60, China). The pH values of AnoxMBBR and AeCSTR were kept at 7.5 and 7.3, respectively. The reactors were continuously operated at hydraulic retention time of 48h. The LFL treatment performance of these reactors was evaluated in terms of COD,  $\text{NH}_4^+$ ,  $\text{NO}_2^-$  and  $\text{NO}_3^-$  removal efficiencies.

### ***Analysis***

Samples were collected from influent and effluent of the AnoxMBBR as well as effluent of the AeCSTR. Then, samples were immediately centrifuged (Eppendorf, Hamburg, Germany) and filtered using cellulose acetate syringe filters with  $0.45\mu\text{m}$  pore sizes (Sartorius AG, Gottingen, Germany). The COD measurements were carried out according to the dichromate-closed reflux Colorimetric Method described by Standard Methods (Standard Methods, 5220 D). The  $\text{NH}_4^+$ ,  $\text{NO}_2^-$  and  $\text{NO}_3^-$  were measured using an ion chromatography (Dionex ICS-3000, Sunnyvale, CA, Japan). The temperature and pH values of both reactors were online measured and recorded in situ daily. In the AeCSTR, DO concentration was measured by a DO meter (Thermo, Orion 4 Star, Indonesia).

## **RESULTS AND DISCUSSIONS**

### ***The COD Removal Performance of AnoxMBBR/AeCSTR***

The A/O system has long been used to remove simultaneous nitrogen and COD from municipal and industrial wastewater. It is known that the COD concentration plays an important role in A/O system, which directly affects the reaction rate of denitrification and nitrification. In this part of the study, the continuous AnoxMBBR/AeCSTR system performance was evaluated in terms of COD removal at HRT of 48h. The variations of COD concentrations in this system are presented in Figure 1. The influent COD concentration throughout this study was average  $10550\pm 350$  mg/L. The COD removal efficiency of both reactors was quite stable. The COD removal was relatively low at AnoxMBBR, corresponding to around 20% removal efficiency and about 8400 mg COD/L effluent concentration.

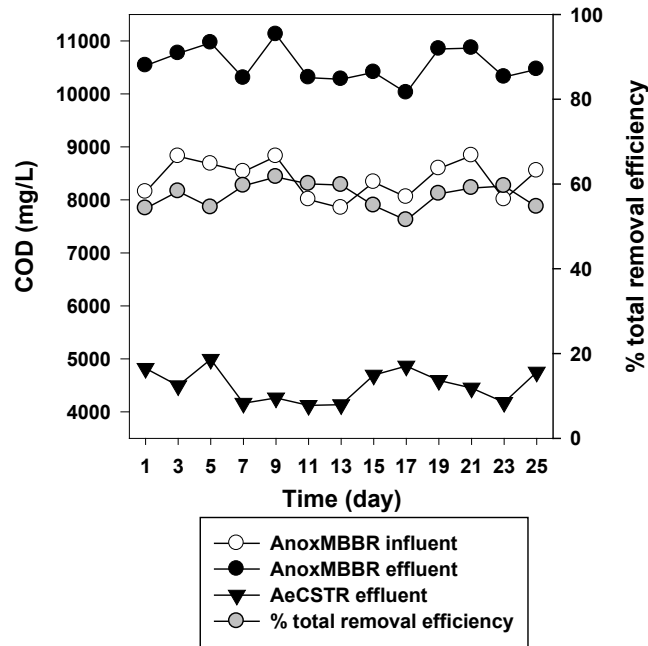


Figure 1. COD Removal Performance of Sequential AnoxMBBR/AeCSTR

The low COD removal efficiency in AnoxMBBR was due to very high influent COD concentration which caused inhibitions on microbial activities of heterotrophic denitrification (Liang et al., 2015). However, the COD concentration under aerobic conditions decreased sharply and reached 4500 mg/L in the AeCSTR. The increase of COD removal efficiency in AeCSTR can be explained with increasing of microbial activity. Additionally, total COD removal efficiency of this system was about 58 %. Liu et al., (2017), investigated the performance of the two-stage A/O combined membrane bioreactor to remove simultaneous COD and nitrogen from LFL. Similar to our results, they reported that the microorganism in Anoxic zone of first stage A/O showed low COD removal performance while the microbial activity increased in aerobic zone.

### The Nitrogen removal performance of AnoxMBBR/AeCSTR

Heterotrophic denitrification process is occurred by reduction-oxidation reactions from nitrate to nitrogen gas via microorganisms under anoxic conditions. The nitrate and organic matter in this process used as electron acceptor and electron donor, respectively. In this study, the influent nitrate concentration was kept constant at 650 mg/L. The denitrification performance of AnoxMBBR during landfill leachate treatment was evaluated in terms of nitrate removal (Figure 2). Similar to COD removal efficiency, the NO<sub>3</sub><sup>-</sup> removal was quite stable and nitrate removal efficiency observed as about 40%, corresponding to effluent nitrate concentration of about 390 mg/L. It seems that the nitrate removal rate depends on COD removal performance. The nitrite accumulation was not also observed in the effluent of AnoxMBBR (Figure 2). Additionally, the ammonium removal during anoxic operation can be negligible at ammonium concentration of 55 mg/L, which was probably used for microbial growth

The ammonium conversion under aerobic conditions is carried out in two steps which oxidized to nitrite by ammonium oxidizing bacteria and nitrate by nitrite oxidizing bacteria, respectively, as shown in the following reactions (Eqs. 1-2). The partial nitrification consists of AOB enrichment and NOB washout while complete nitrification consists of NOB enrichment.



The performance of AeCSTR during landfill leachate treatment using AnoxMBBR/AeCSTR was evaluated in terms of the comple/partial nitrification. AeCSTR was operated at HRT of 48 h and DO concentration over 4mg/L.

Figure 3 shows the ammonium conversion performance of the AeCSTR. The influent  $\text{NH}_4^+$  concentration was about  $1620 \pm 75$  mg/L throughout AeCSTR operation.

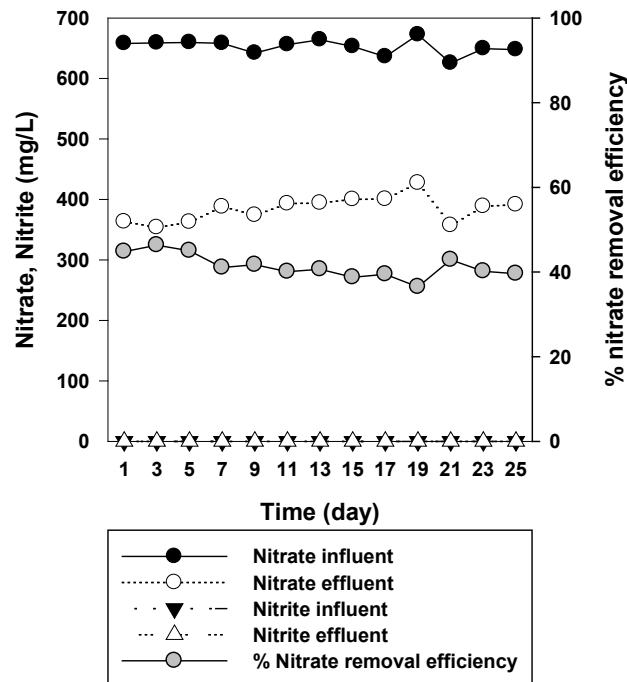


Figure 2. The Nitrate and Nitrite Removal Performance of AnoxMBBR

The  $\text{NH}_4^+$  concentrations decreased sharply in AeCSTR, corresponding to 71.5% removal efficiency. Additionally, effluent  $\text{NH}_4^+$  concentrations were approximately 463 mg/L. The nitrite accumulation under aerobic conditions was clearly observed and effluent nitrite and nitrate concentrations were 1580 mg/L  $\text{NO}_2^-$  and 350 mg/L  $\text{NO}_3^-$ , respectively. This indicated that partial nitrification carried out in the reactor. This result was similar that of previous study (Fitzgerald et al., 2015), in which the AOB population during high  $\text{NH}_4^+$  oxidation was found to dominant.

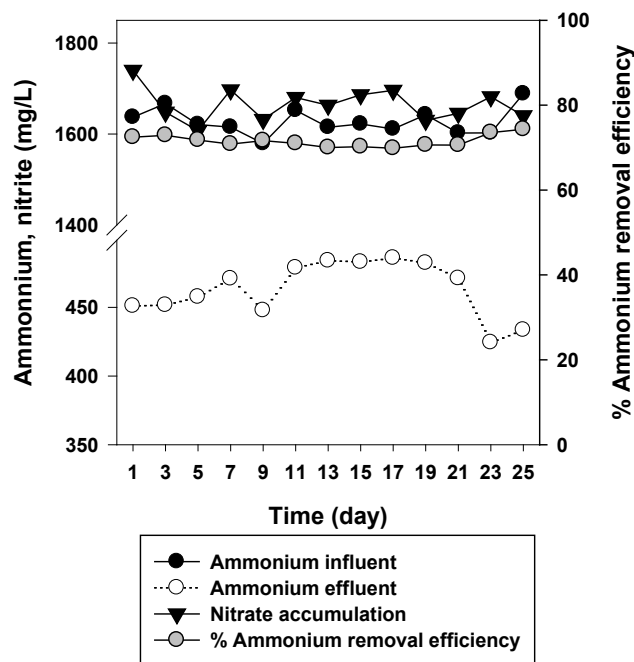


Figure 3. The Ammonium Removal and Nitrite Accumulation Performance of AeCSTR

## CONCLUSION

The following conclusions from this study can be drawn:

- The denitrification efficiency of raw LFL in the AnoxMBBR were ineffective due to high COD concentration in influent wastewater
- In AeCSTR, the partial nitrification was observed at HRT of 48-h.
- The AnoxMBBR/AeCSTR system 57% and 74% COD and ammonium removal efficiencies were obtained, respectively.
- This study demonstrates that the AnoxMBBR/AeCSTR system is in favor of LFL treatment.
- However, this study demonstrates that this system still remained insufficient to meet discharge standards and integrated systems are required.

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