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Anoxic/aerobic granular active carbon assisted MBR integrated with nanofiltration and reverse osmosis for advanced treatment of municipal landfill leachate

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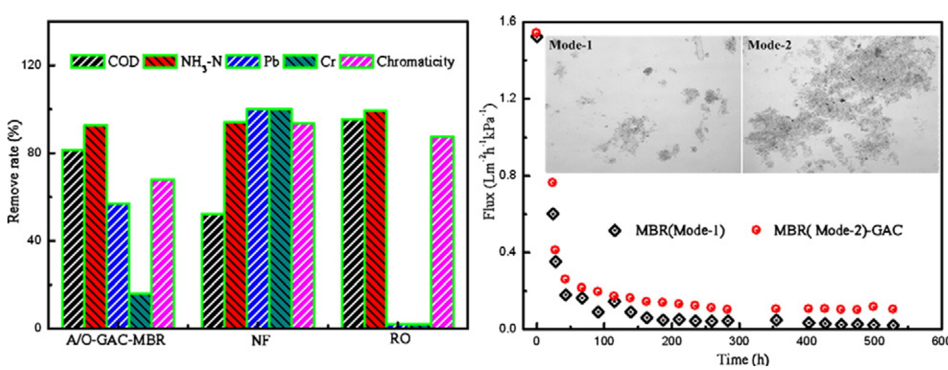
HIGHLIGHTS

- Anoxic/aerobic granular active carbon assisted MBR was used for leachate treatment.
- Removal efficiency of high-valence metals was more than low-valance ones.
- Granular active carbon enhanced flocculation and alleviated membrane fouling.
- The color of MBR effluent decreased from 125 to 8 times after NF treatment.
- RO effluent can meet the requirement of industrial reutilization very well.

GRAPHICAL ABSTRACT



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ABSTRACT

Anoxic/aerobic granular active carbon assisted membrane bioreactors (A/O-GAC-MBR) integrated with nanofiltration (NF)–reverse osmosis (RO) were used for leachate treatment of the Taizhou Municipal Landfill plant. In order to investigate the effect of GAC on the performance of MBR, a hybrid MBR with GAC dosed and a conventional MBR were operated in parallel. Two pilot scale MBRs showed excellent and stable removal efficiency with average above 80% for chemical oxygen demand (COD) and ammonia-nitrogen (NH_3-N). It was noticed that the amount of high-valence metal ions were removed more than low-valence ones. By comparison, the addition of GAC not only improved the removal of hazardous organic pollutants and heavy metals, but also enhanced bioflocculation and particle size of flocs, which greatly alleviated membrane fouling. NF and RO membranes were then used for advanced treatment of MBR effluents. NF membrane exhibited excellent color removal rate of 93.75% compared to 41.82% of A/O-MBR. With the advantage of NF pretreatment, RO membrane held a steadier and higher flux as well as salt rejection than other cases reported. The final permeate from RO was proved to exceed related qualification for reutilization and can easily be recycled in industry.

Abbreviations: A/O-MBR, anoxic/aerobic membrane bioreactor; NF, nanofiltration; RO, reverse osmosis; GAC, granular activated carbon; COD, chemical oxygen demand; NH_3-N, ammonia-nitrogen; MSW, municipal solid waste; PVDF, polyvinylidene fluoride; HRT, hydraulic retention time; MLSS, mixed liquor suspended solid; SVI, sludge volume index; SEM, scanning electron microscope; R_m, inherent resistance of the membrane (m^{-1}); R_c, resistance of cake layer (m^{-1}); R_{if}, resistance of membrane organic fouling (m^{-1}); R_{of}, resistance of membrane organic fouling (m^{-1}); R_t, total resistance (m^{-1}); J_w, permeate flux of membrane ($L \cdot m^{-2} h^{-1}$); M, permeate viscosity ($Pa \cdot s$); ΔP, transmembrane pressure (kPa); M, compressed coefficient (m); m′, attenuation coefficient; J_tm, flux at t moment ($L \cdot h^{-1} m^{-2}$); J_i, flux at i moment ($L \cdot h^{-1} m^{-2}$).

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1. Introduction

In China, more than 150 million tons of municipal solid wastes (MSW) are produced with an increase rate of 8%–10% every year, and approximately 80% MSW are treated by local landfills [1]. The major problem caused by landfills is the generation of toxic and hazardous leachates. The leachate treated by conventional methods is hard to be allowed to discharge because it often contains lots of hazardous organic contaminants, ammonia, heavy metals, and inorganic salts [2]. Therefore, the development of new technologies for the advanced treatment of leachate has attracted more and more attention.

For the past few years, various methods such as air stripping, activated carbon adsorption, coagulation–flocculation, chemical oxidation, membrane separation and sequencing batch reactors (SBR) have been applied for the landfill leachate treatment [3–5]. Among these technologies, the biological process has been expected to be an effective method to treat landfill leachate due to its good performance, simplicity, and low cost [6]. Membrane bioreactor (MBR), as one of the most promising biological technologies, has big potential for the stable and efficient biological treatment of leachate [7–9]. However, many previous practices have also demonstrated that the biological methods are only effective to treat younger leachate (<1–2 years old) with high BOD_5 [10]. While applying to treat the older ones (>10 years old), there are several problems and challenges presented, which was mainly attributed to the characteristics of leachate such as low BOD_5/COD, high concentration of COD and NH_3-N, and the existence of toxic heavy metals. Moreover, membrane fouling has always been addressed as a major problem limiting the wide application of MBR for landfill leachate treatment [11]. To overcome these difficulties and comply with the increasingly stringent regulation such as the improved standard of GB 16889-2008, more efficient and reliable technology should be required eagerly. As reported, almost no single process seems available for efficient and simple leachate treatment. Hence, the combined process like the hybrid physical/chemical–biological process is necessary to be proposed to deal with these problems [12,13].

Advanced chemical oxidations such as ozonation and electrolysis placed before MBR process could enhance the removal efficiency of recalcitrant substances during the old landfill leachate treatment [14,15]. However, these chemical oxidation technologies may be hard to be widely applied in the industry due to their high cost and complicated operation in treatment [16]. Comparatively, granular active carbon (GAC) as the pretreatment unit or additives integrated with biological process such as sequencing batch reactor and activated sludge has been widely used for the landfill leachate treatment due to its low-cost, high removal efficiency and easy to operation [17,18]. Moreover, recent researches have revealed that the addition of GAC into mixed liquid was an effective route to alleviate membrane fouling [19,20]; from our previous experience, it might be beneficial to combine GAC with MBR process for landfill leachate treatment. Furthermore, in order to meet the higher requirement for reutilization of leachate, the pressure-driven reverse osmosis (RO) and nanofiltration (NF) membrane technology seems to be an effective alternative for further desalination and purification. As reported, a remarkably high removal of COD and NH_3-N were achieved by using a two-stage process integrating MBR with a post-treatment of RO for old leachate treatment [21], however, the severe membrane fouling that happened was addressed as a major problem limiting the long time operation of RO. Complementary combination of NF placed before RO appears to be a cost-effective approach in comparison with the other methods, which are commonly used at present and can effectively remove the annoying membrane foulants such as small molecular organic matter.

Based on the above observation, the A/O-GAC-MBR system integrated with nanofiltration (NF) and RO dual membrane systems for old leachate treatment with a pilot scale set up was evaluated for the project design of a 1000 m^3/d landfill leachate treatment plant in Taizhou. The anoxic biological system was built as pretreatment to effectively remove refractory polycyclic and heterocyclic organics, and then the aerobic MBR process was supplied with stable and efficient degradation of organic matter and nitrogen. The main interests were taken in exploring the effect of GAC dosage on removal performance, properties of biomass and membrane filtration. Finally, the NF and RO dual membrane systems were applied to provide advanced post-treatment of leachate. The performance of each unit of COD, NH_3-N, and heavy metal removal was investigated in detail. The membrane fouling of MBR, NF, and RO processes was also emphasized.

2. Materials and methods

2.1. Landfill leachate samples

Raw leachate wastewater was collected from the Taizhou Landfill Plant. The chemical physical characteristics of landfill leachate are listed in Table 1. Clearly, the BOD_5/COD ratio was lower than 0.14 and hence it was classified as an old and non-biodegradable leachate.

2.2. Experiment setup

The pilot scale experimental setup is illustrated in Fig. 1. In order to analyze the impact of additional GAC on the leachate treatment and membrane fouling, two identical-scale MBRs, one conventional MBR (Mode-1) and another granular active carbon assisted MBR (Mode-2,) were run in parallel under the same conditions. The bioreactors consisted of an anoxic bioreactor (50 L) and an aerobic bioreactor (50 L) with an immersed polyvinylidene fluoride (PVDF) hollow fiber membrane module (pore size 0.2 μm, effective membrane area 0.25 m^2). Raw leachate was fed from a storage tank to anoxic bioreactor by a diaphragm pump, and then the effluent was pumped to the aerobic bioreactor. Two MBRs were operated under constant flux (6.25 L/m^2 h) mode with 10 min suction followed by 5 min relaxations. Seed sludge in bioreactors was obtained from the local municipal wastewater treatment plant, and the mixed liquor suspended solid (MLSS) of activated sludge was set at 4–5 g/L throughout the whole process. The solids retention time (SRT) and hydraulic retention time (HRT) were 40 and 2 days for both anoxic and aerobic bioreactors, respectively. Air was supplied continuously to the aerobic bioreactor at a constant flow rate in order to provide dissolved oxygen (DO, 2–4 mg/L) and alleviate membrane fouling. The transmembrane pressure (TMP), as an indicator of the extent of membrane fouling, was continually monitored.

NF/RO experiments were carried out at a controlled temperature of 25 °C. To obtain adequate pressure, a high-pressure pump was used to feed MBR effluent into the cross flow NF modules with DL plate-frame membranes (Osmonics, the effective membrane area 66 cm^2). After magnification treatment, the NF effluent was fed into the cross flow RO modules with BW plate–frame membranes (Filmtech, the effective membrane area, 66 cm^2). NF and RO were carried out at an operating pressure of 1.4 and 2.8 MPa, respectively. The concentration of NF and RO was then mixed with secondary effluent and returned to the feed tank to keep the mass balance.

2.3. Membrane cleaning in MBR

The cleaning of membrane module was carried out periodically by physical and chemical methods. First, the membrane was washed by pure water to remove the cake layer attached on its surface, and then a chemical cleaning process (cleaned by the mixture of 0.2% hydrochloric acid and 0.8% citric acid for 4 h, and then cleaned by 1% NaOH solution and 0.2% NaClO solution for 16 h) was conducted. Meanwhile, the permeate flux of cleaned membrane in pure water was tested.