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Oscillating membrane photoreactor combined with salt-tolerated *Chlorella pyrenoidosa* for landfill leachates treatment

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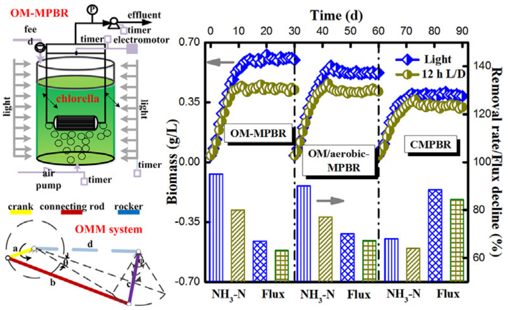
GRAPHICAL ABSTRACT



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ABSTRACT

In this study, a novel oscillating membrane (OM) photoreactor combined with salt-tolerated *Chlorella* was developed for old landfill leachates treatment, in which harvesting of highly-active algae was easily performed on large scale. Compared with control membrane photo-bioreactor (CMPBR), OM-MPBR exhibited excellent NH_3-N removal efficiency as high as 94.0%. With light time prolonged, an increase in biomass production and NH_3 removal rates was observed due to more energy provided for *Chlorella* cells. By comparison, it was found the highest membrane flux (99.6 L/m^2 h bar) was obtained in OM-MPBR, which was attributed to strong shear stress on interface of liquid/membrane effectively reducing bio-foulants. It was clear that energy consumptions of OM-MPBR on biomass productivity (0.68 kWh/kg cell) and NH_3 removal (0.0151 kWh/kg NH_3-N) were lower than

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Abbreviations: A_n, membrane area needed (m^2); A_{ref}, membrane area of the referenced municipal MBR installation (m^2); C_A, energy consumption for compressing the air of full-scale MBR installation (kWh/m^3); *CIP*, energy consumption for unit operations related to membrane area membrane (kWh/m^3); *CMPBR*, conventional membrane photo-bioreactor; *COD*, chemical oxygen demand (mg/L); *DO*, dissolved oxygen (mg/L); E_v, estimated energy consumption based on permeate volume (kWh/m^3); E_w, estimated energy consumption based on dry weight of harvested biomass (kWh/kg); *EPS*, extracellular polymeric substance; *HRT0*, hydraulic retention time (h); J_p, permeate flux (L/m^2 h); L_p, permeability (L/m^2 h kPa); NH_3-*N*, ammonia-nitrogen (mg/L); OD_p, the optical density of the permeate; *OM-MPBR*, oscillating module membrane photo-bioreactor; *OM/aerobic-MPBR*, oscillating module/aerobic membrane photo-bioreactor; P_P, energy consumption for permeate pumping of full-scale MBR installation (kWh/m^3); *PVDF*, polyvinylidene fluoride; R_c, cake resistance (m^{-1}); *Re*, Reynolds number; R_f, membrane fouling resistance (m^{-1}); R_{if}, inorganic fouling resistance (m^{-1}); R_m, initial clean membrane resistance (m^{-1}); R_{of}, organic fouling resistance (m^{-1}); R_t, total membrane filtration resistance (m^{-1}); R_i, irreversible membrane fouling; *SEM*, scanning electron microscope; *SRT*, solid retention time (day); *TMP*, transmembrane pressure (kPa); *v*, cross flow velocity (m/s); Δp_{TM}, transmembrane pressure (bar); η, plastic viscosity (Pa·s); μ, specific growth rate (day^{-1}); r_A, Ratio of membrane area needed (A_n) to the referenced municipal MBR area (A_{ref}); ρ, solid concentration of bacteria in the bioreactor (kg/m^3); η_a, harvesting efficiency of photosynthetic bacteria biomass (%)

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CMPBR. The new coupling system opens a door to scalable development of promising and economical MBR for environmental pollution control and biomass energy production.

1. Introduction

With the increase of population density and social economy, million tons of municipal solid wastes (MSW) are produced in China per year (Liu et al., 2006). Although landfilling is considered as the most common method used for the disposal of MSW in most countries (Kjeldsen et al., 2002; Dong et al., 2014), it often generates large amounts of landfill leachates after the decomposition of MSW, which is always regarded as a kind of refractory wastewater (Sniffen et al., 2017). Leachates with pungent odor and dark color contains high content of toxic organic pollutants, ammonium (NH_3-N), heavy metals and a wide range of complicated compositions. The discharge of leachates without effective treatment may cause serious pollution to the groundwater aquifers and pose most significant hazards to human health (Wang et al., 2016; Wu et al., 2018; Holloway et al., 2014). Therefore, it is crucial for the development of environmental and effective technology in landfill leachate treatment. To date, various methods such as physical (air striping, multi-stage filtration), chemical (coagulation–flocculation, chemical oxidation), and biological (activated sludge, sequencing batch reactors) processes have been attempted to treat landfill leachate (Pirbazari et al., 1996; Ahmed and Lan, 2012; Hasar et al., 2009; Foo and Hameed, 2009). However, these methods often take disadvantages of high operation cost, low removal efficiency, secondary pollution and requirement of extra additives. Comparatively, membrane bioreactor (MBR) is one of the most promising biotechnology and has great potential for stable and efficient treatment of leachate. Previous studies have shown that simple MBR system can be effective to treat younger leachates (Wang et al., 2014). But for old landfill leachates, high ammoniacal-N, salinity and toxicity still make the wastewater treatment be one of the most bothersome issues. This is mainly due to the fact that the microbial activity and metabolism of activated sludge is severely inhibited by the existence of large numbers of NH_3-N and the soluble inorganic salts which usually disturb the cell osmotic pressure (Lin et al., 2007; Uygur and Kargı, 2004). Hence, development of novel MBR technology will be very essential for solving these problems of environmental pollution.

In the design of novel MBR systems, mitigation of membrane fouling is deemed as the most critical point for long-term stable operation in application. Unlike other sources of fouling, biofouling with self-replication property is difficult to be eliminated by pretreatment, which often causes serious damages to membrane materials (Bilad et al., 2013; Shi et al., 2012). Over past decades, some efforts have been devoted to designing various methods for reducing membrane fouling such as membrane modification (He et al., 2009; Yu et al., 2006), microorganism immobilization (Kim et al., 2015; Lu et al., 2017), operating conditions optimization (Huang et al., 2017), novel component design (Rattananurak et al., 2015) and multiple technologies combination (Zhang et al., 2013, 2014; Qin et al., 2012). In our previous studies, several ways have been tried to move forward, for example, the design of dynamic membrane module as a promising alternative (Qin et al., 2015, 2017). In comparison with air bubbling of conventional MBRs, the vibration of membrane module can create stronger shear stress on the liquid-membrane interface and effectively mitigate the formation of biofilm. Therefore, the attempt on dynamic membrane module will provide more efficient approaches in controlling membrane fouling, which is very important for the development of advanced MBR systems.

Considering the high concentration and complexity of pollutants in old landfill leachates, the screening and cultivation of salt-tolerated high efficient bacteria instead of activated sludge for treating non-degradable wastewater tends to be a good choice (Nguyen et al., 2018). As kind of biomaterials in high productivity, microalgae have been successfully applied in water and wastewater treatment. The microalgae system showed good recycle ability and renewable bioenergy production (Cheah et al., 2016; Angenent et al., 2004). Moreover, the harvested microalgae biomass can also be used as good source of high-value product, animal food and agrochemical applications (Safi et al., 2014; Katiyar et al., 2017). However, it is very difficult to implement the rapid harvesting of algae on a large-scale because of the slow growth rate and poor settling ability (Xu et al., 2014). Although MBR combined with algae has been explored in wastewater treatment and presented good performance in harvesting (Frappart et al., 2011), how to reduce membrane biofouling especially when operated in high biomass is still a big challenge (Meng et al., 2017). Based on these observations, we go forward to think, if we can select some efficient microorganisms, like *Chlorella*, and combine it with dynamic membrane system in old landfill leachate treatment, attractive characteristics including both anti-fouling and harvesting may be obtained.

In this study, a novel oscillating membrane photoreactor combined with salt-tolerated *Chlorella pyrenoidosa* has been developed, which showed higher tolerance in harsh environmental stresses, effective nitrogen removal and excellent anti-fouling performance for old landfill leachates treatment. Compared with our the previously reported oscillating system (Qin et al., 2017), the new designed crank rocker structure of oscillating membrane module can reduce energy consumption and enhance transmission power due to the advantages of simple handiness, low frictional resistance and easy control on swing angles. To the best of our knowledge, such algae/membrane photo-bioreactor with OM (OM-MPBR) for integrating the microbe's cultivation together with high salinity landfill leachate treatment has not been reported up to now. The main interests were taken in designing the new *Chlorella*/OM-MPBR system and evaluating their performance including biomass production, nutrient removal efficiency, membrane flux and energy consumption. In order to further improve biomass production and reduce operating costs, the effects of illumination time and aeration on MPBRs were also investigated.

2. Materials and methods

2.1. Cultivation of Chlorella pyrenoidosa

Chlorella pyrenoidosa (FACHB-28) powders (Chinese Academy of Sciences, China) were firstly cultivated in 500 ml flasks with the sterile BG11 medium (including 40 mg/L $K_2HPO_4\cdot3H_2O$, 36 mg/L $CaCl_2\cdot2H_2O$, 75 mg/L $MgSO_4\cdot7H_2O$, 6 mg/L ferric ammonium citrate, 20 mg/L Na_2CO_3, 6 mg/L citric acid, 1 mg/L EDTA). During the large-scale culture, the intensity of light irradiation, temperature, pH and aeration rate were adjusted at about 3000 Lux, 28 °C, 7.0 and 2.5 L/min, respectively. The growth of *Chlorella pyrenoidosa* in BG11 culture at different initial cell concentrations was shown (Fig. S1a). Obviously, the growth trend of *Chlorella pyrenoidosa* at different initial concentrations was similar. Secondly, the cultivated *Chlorella* microbes were acclimated according to the character of old landfill leachates. The initial cell absorbance $OD_{625} = 0.15$ (DCW = 0.037 g/L) was used in the different MPBR systems.

2.2. Landfill leachate samples

Actual leachates were collected from Taizhou Landfill Plant (salinity of 7%, average NH_3-N of 230 mg/L and COD of 304 mg/L), which was