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A novel forward osmosis system in landfill leachate treatment for removing polycyclic aromatic hydrocarbons and for direct fertigation

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HIGHLIGHTS

- A novel FO system using NH_4HCO_3 as the DS was investigated.
- The FO system can effectively eliminate PAHs pollutants in landfill leachate.
- The diluted DS can be directly used as a liquid fertilizer.
- Landfill leachate treated by FO had no toxicity effects on zebrafish and human cells.

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ABSTRACT

Landfill leachate (LL) is harmful to aquatic environment because it contains high concentrations of dissolved organic matter, inorganic components, heavy metals, and other xenobiotics. Thus, the remediation of LL is crucial for environmental conservation. Here, a potential application of the forward osmosis (FO) filtration process with ammonium bicarbonate (NH_4HCO_3) as a draw solution (DS) was investigated to remediate membrane bioreactor-treated LL (M-LL). After the leachate treatment, the toxicity and removal efficiencies of polycyclic aromatic hydrocarbons (PAHs) were evaluated using zebrafish and cultured human cells. The water recovery rate was improved using the current protocol up to 86.6% and 91.6% by both the pressure retarded osmosis (PRO) mode and the forward osmosis (FO) mode. Water flux increased with the increasing DS concentrations, but solution velocities decreased with the operation time. Toxicity tests revealed that the M-LL treated by NH_4HCO_3 had no toxic effect on zebrafish and human cells. Moreover, green fluorescent protein (GFP) expression in the transgenic zebrafish *Tg(cyp1a:gfp)* induced by PAHs was very weak compared to the effects induced by untreated M-LL. Since the diluted DS met local safety requirements of liquid fertilizer, it could be directly applied as the liquid fertilizer for fertigation. In conclusion, this novel FO system using NH_4HCO_3 as the DS provides a cheap and efficient protocol to effectively remove PAHs and other pollutants in LL, and the diluted DS can be directly applied to crops as a liquid fertilizer, indicating that this technique is effective and eco-friendly for the treatment of different types of LL.

1. Introduction

Remediation of landfill leachate (LL) is quite challenging, as it contains large amount of organic matter, heavy metals, and inorganic salts, which lead to high levels of chemical oxygen demand (COD), biochemical oxygen demand (BOD), ammonia-nitrogen, and wastewater pigmentation (Bohdziewicz et al., 2008; Fernandes

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et al., 2014; Ricordel and Djelal, 2014). Because of the heterogeneous composition, abundant anthropogenic chemicals, and neglected environmental regulation, LL poses elevated risks to ground and surface water resources. Leachate contamination into the soil, groundwater, surface water, and air can not only affect the surroundings but also disturb the ecological balance (Cortés-Lorenzo et al., 2014). Among LL contaminants, polycyclic aromatic hydrocarbons (PAHs) and other organic materials can induce carcinogenic and mutagenic effects in both human and aquatic organisms (Asakura et al., 2004), and thus they have been listed as priority pollutants by the US Environmental Protection Agency (US-EPA) (Chen and White, 2004). The presence of retained pollutants in LL, especially PAHs, implies that LL has itself become one of the most dangerous secondary environmental pollutants (Widziewicz et al., 2012). Therefore, it is critical to explore an effective and economical way to treat LL.

Coagulation, sedimentation, activated carbon adsorption, and membrane technology are the commonly used primary techniques for LL treatment. Advanced treatments such as photo-catalytic oxidation, electrochemistry, and activated sludge are proved effective in the treatment of organic matter, heavy metals, and ammonia nitrogen (Abood et al., 2013; Singh et al., 2014), but they are not effective in removing total soluble solids. Moreover, they possess multiple other drawbacks such as unstable treatment effects, a large occupancy area, high energy consumption, and cumbersome processing (Singh et al., 2012). Membrane-based techniques, such as microfiltration, ultrafiltration, nanofiltration, and reverse osmosis (RO) are commonly used for wastewater treatment (Peters, 1998; Ince et al., 2010; Wang et al., 2014a), but energy consumption is very high. The previous LL treatment process in the Changsheng Bridge Landfill (Nanan District, Chongqing, China) included six common processes: pretreatment, upflow anaerobic sludge blanket, anaerobic bioreactor, aerobic bioreactor, membrane biological reactor (MBR) and RO. The RO process itself has several drawbacks such as: (1) low recovery rates (<70%); (2) high energy consumption and operation cost (about $ 7.5/ton); and (3) high investment cost (about $1200/year). To decrease costs and enhance recovery, we explored the feasibility of replacing RO with forward osmosis (FO). FO is an alternative membrane process, which offers many advantages such as lower fouling potential, less energy consumption, simplicity, and higher recovery (Cath et al., 2006, 2010; Mi and Elimelech, 2010; Zhao et al., 2012; Valladares Linares et al., 2013; Werner et al., 2013). Using the natural phenomenon of osmosis, it saves time and money compared to other membrane techniques. Currently, FO is widely used in seawater desalination, water purification, wastewater treatment, food and pharmaceutical processing, etc. (McCutcheon et al., 2006; Cornelissen et al., 2008; Kong et al., 2014; Wang et al., 2014b; Zhang et al., 2014).

Besides membrane fouling, selecting optimal draw solution (DS) components and conditions are the main constraints in the practical application of the FO technique. Air scouring and chemical cleaning can eliminate reversible and irreversible membrane fouling. To maintain the concentration of the DS, the addition of concentrated salt solutions is highly recommended (Dong et al., 2014). The abilities to regenerate the concentrated DS and dispose effluents are perhaps the most important criteria for selecting a DS. Inorganic compounds (NaCl, $MgCl_2$, and $A1_2(SO_4)_3$) and organic compounds (glucose, fructose, and alcohols) are quite commonly used as DSs, but each of them has clear drawbacks (Achilli et al., 2010; Li et al., 2011; Su et al., 2012; Sato et al., 2014). Recently, different DSs were considered including zwitterions, polyacrylic acid sodium salt, fertilizer, and magnetic protein (Ge et al., 2012; Chanukya et al., 2013; Lutchmiah et al., 2014).

Interestingly, when a fertilizer like NH_4HCO_3 was used for FO desalination, the extracted diluted DS could be directly applied for fertigation (Phuntsho et al., 2011). This option offers the advantage that the diluted DS of NH_4HCO_3 becomes a safe and readily available fertilizer without desalination and at decreased cost. The purposes of the current study were twofold: to carry out a preliminary assessment for the feasibility of using NH_4HCO_3 as a DS and a fertilizer after a FO-based M-LL treatment in Changsheng Bridge Landfill (Nanan District, Chongqing, China) and to determine whether the effluents met safety assessments for subsequent use in irrigation.

Generally, BOD, COD, and total organic carbon (TOC) are the major physicochemical parameters to evaluate wastewater quality. However, these physicochemical parameters cannot evaluate acute toxicity and genotoxic hazards to aquatic organisms caused by wastewater (Ma et al., 2005; Niu et al., 2016). Fortunately, aquatic organisms can be used to evaluate the toxic effects of the post-treated wastewater and assess the detoxification efficiencies of multiple processes (Ma et al., 2005). Previous reports demonstrated that zebrafish (*Danio rerio*) provide a suitable model organism to explore DNA damage and toxicology, because zebrafish larvae are sensitive to poisonous substances (Pei and Strauss, 2013; Dai et al., 2014). Bioassays with wild-type zebrafish and transgenic zebrafish have been proved to be a faster and more cost-effective way for toxicity testing of pollutants in environmental samples (Carvan et al., 2000; Carlsson et al., 2009; Kanungo et al., 2011). In addition, the cytochrome P450 1A (Cyp1a) gene is frequently upregulated by PAHs through activating the aryl hydrocarbon receptor (AhR) pathway (Kim et al., 2013). Thus, the transgenic zebrafish *Tg(cyp1a:gfp)* can be used for monitoring PAHs in water.

In this study, a FO membrane system with NH_4HCO_3 as the DS was used to treat M-LL. Different concentrations of the DS, solution velocities, and membrane orientations were examined. Water recovery rates of M-LL were measured, and the feasibility of applying the diluted DS for direct fertigation was explored. Finally, wild-type zebrafish and cultured human cells were used as biological models to test the toxicity of posttreated M-LL, while transgenic zebrafish *Tg(cyp1a:gfp)* were used to quantify the detoxification of PAHs. Our results revealed that this novel FO system with NH_4HCO_3 as the DS is not only effective and eco-friendly for the LL treatment and direct fertigation, but also can eliminate various pollutants in the LL, including PAHs.

2. Materials and methods

2.1. Ethics statement

All experiments involving zebrafish conformed to the “Guide for the Care and Use of Laboratory Animals” (Eighth Edition, 2011. ILARCLS, National Research Council, Washington, D.C.). The animal protocol was approved by the Institutional Animal Care and Use Committee of Chongqing Institute of Green and Intelligent Technology, Chinese Academy of Sciences (Approval ID: ZKCQY0069).

2.2. Solutions

Ammonium bicarbonate, hydrochloric acid, and sodium hydroxide were purchased from KeLong Chemical, Chengdu, China. All chemicals were of analytical grade. The feed solution used in this experiment was MBR-treated LL, collected from Changsheng Bridge Landfill. Water quality measurements are shown in Table S1.

2.3. Forward osmosis membrane

The FO membrane was thin film composite (TFC) polyamide membrane (Hydration Technologies Innovation, Albany, USA). This