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Using nano-attapulgite clay compounded hydrophilic urethane foams (AT/HUFs) as biofilm support enhances oil-refinery wastewater treatment in a biofilm membrane bioreactor

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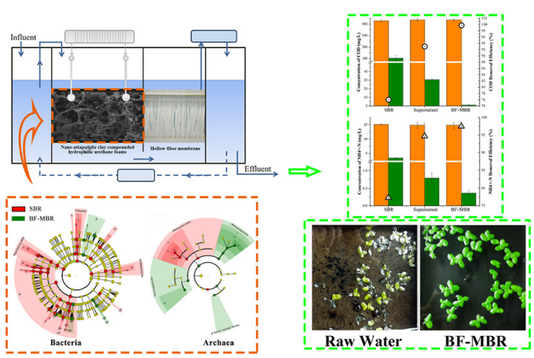
HIGHLIGHTS

- Refractory petroleum refinery wastewater was treated using BF-MBR with AT/HUFs as a biofilm support.
- High COD, NH_4^+ and hazardous material removal rates were achieved.
- Giant duckweeds grew well in BF-MBR-treated wastewater.
- Efficient degraders, bacteria and archaea on AT/HUF carriers were enriched.

GRAPHICAL ABSTRACT



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ABSTRACT

Petroleum refinery wastewater (PRW) treatments based on biofilm membrane bioreactor (BF-MBR) technology is an ideal approach and biofilm supporting material is a critical factor. In this study, BF-MBR with nano-attapulgite clay compounded hydrophilic urethane foams (AT/HUFs) as a biofilm support was used to treat PRW with a hydraulic retention time of 5 h. The removal rate of 500 mg/L chemical oxygen demand (COD), 15 mg/L NH_4^+ and 180 NTU of turbidity were 99.73%, 97.48% and 99.99%, which were 23%, 20%, and 6% higher than in the control bioreactor, respectively. These results were comparatively higher than that observed for the sequencing batch reactor (SBR). The death rate of the *Spirodela polyrrhiza* (L.) irrigated with BF-MBR-treated water was 4.44%, which was similar to that of the plants irrigated with tap water (3.33%) and SBR-treated water (5.56%), but significantly lower than that irrigated with raw water (84.44%). The counts demonstrated by qPCR for total bacteria, denitrifiers, nitrite oxidizing bacteria, ammonia oxidizing bacteria, and ammonia-oxidizing archaea were also higher in BF-MBR than those obtained by SBR. Moreover, the results of 16 s rRNA sequencing have demonstrated that the wastewater remediation microbes were enriched in AT/HUFs, *e.g.*,

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urethane foam (AT/HUF)
Microbiome

Acidovorax can degrade polycyclic aromatic hydrocarbons, and *Sulfuritalea* is an efficient nitrite degrader. In summary, BF-MBR using AT/HUF as a biofilm support improves microbiome of the actived sludge and is reliable for oil-refinery wastewater treatment.

1. Introduction

Petroleum refinery wastewater (PRW) contains a wide range of petroleum hydrocarbons, heavy metals, aromatic compounds, phenolic substances, and other noxious compounds that are generated during the petroleum refining and manufacturing processes (Cappello et al., 2016). If PRW is not appropriately treated or recycled, their by-products can cause serious threats to the environmental microbiome, living organisms, and aquatic ecosystem (Huang et al., 2016; Yang et al., 1997; Yu et al., 2017). When amounts and types of PRW products increase, the treatment of such an anoxic-oxic sludge becomes difficult. (Ma et al., 2009). Therefore, it is crucial to degrade or recycle these pollutants in the interest of environmental safety. For this purpose, physical, chemical, and biological processes have been developed to remediate oil refinery wastewater (Pinzon Pardo et al., 2007; Yang et al., 2015), in which biological treatments are well established (Pinzon Pardo et al., 2007). These biological treatments are considered as important and low-cost methods for sewage management that includes the activated sludge process, anoxic-oxic (A/O) process, fluidized bed reactors, membrane bioreactors (MBRs), and the biofilm process (Guo et al., 2009). However, the high concentration of chemicals in wastewater often affects the growth and metabolism of microorganisms (Liu et al., 2014), limiting the efficacy of biological methods, which is still a challenging issue regarding their development.

Over the last few decades, MBRs have become an established operational technology for domestic, industrial, municipal and oil refinery wastewater treatment, along with its application in conservation and recycling (Iorhemen et al., 2016). The MBR technology offer advantages over all the conventional wastewater treatment systems that use activated sludge (Sun et al., 2014), which include the exceptional effluent quality, low sludge production, and high biodegradation efficiency (Cai et al., 2016; Zhang et al., 2017). However, membrane fouling phenomenon have become a major obstacle to the sustainability of MBRs for wastewater treatments (Ivanovic and Leiknes, 2012). Although, there is no clear agreement to tackle the negative impact on MBR technology, addition of organic and inorganic polyelectrolytes (Subtil et al., 2014), powdered activated carbon (PAC) (Remy et al., 2010), biopolymer (Koseoglu et al., 2008), and salt metals (Zhang et al., 2011), could be preferred as this would enhance the efficiency of the MBR operating system. The contaminant removal ability and efficiency are affected by the microorganisms and the method of immobilization. In principle, numerous materials could be used as a biofilm support, however, only a few are commercially applied in full-scale systems, such as cord media, RBC media, sponge and plastic media and granular activated carbon (GAC). Porous support-based carrier binding method immobilized microorganism present in most practical applications, the flexible polyurethane foam-type material into the most widely used method of immobilized microorganism support (Ivanovic and Leiknes, 2012). The biofilm membrane bioreactor (BF-MBR) is a type of advanced MBR in which a combination of a biofilm and MBR processes is used that could prove helpful in overcoming some limitations of MBR technology (Ivanovic and Leiknes, 2008), caused as a result of attribution of more extra-cellular polymeric substances (EPS) released into the bulk suspension (Yang et al., 2009). However, the effects of biofilm formation on MBR are still far away from consensus. They normally provide a higher filling fraction and surface area for biofilm growth, which can lead to an increased performance of bioreactor. Whether the use of such type of carriers is not consensus in the long-term biofilm is still unclear. This is due to the lower rate of fouling in the biofilm-MBR than that in the MBR alone with a comparable removal efficiency of organic and nitrogenous pollutants (Sun et al., 2015). Compared to the traditional MBR, the biofilm grown on carriers can reduce the concentration of suspended degraders, membrane fouling, and more efficient to degrade pollutants. In addition, BF-MBR can be operated with higher fluxes and are more compact to control membrane fouling (Zhang et al., 2017). Thus, to develop a novel or advanced BF-MBR operating system to treat PRW in a better way is needed.

In this study, using nano-attapulgite clay compounded hydrophilic urethane foam (AT/HUF) as the biofilm carrier, a novel BF-MBR was set up. Particularly, the focus was to develop a recombinant technology to improve the efficiency of oil-refinery wastewater treatment in comparison to the sequencing batch bioreactor (SBR). The mechanisms responsible for an improved efficiency and the microbial communities have also been investigated using the 16S rRNA sequencing technique and qPCR.

2. Materials and methods

2.1. Physicochemical characteristics analysis of water and activated sludge

The activated sludge and wastewater were stockpiled from an aeration tank of the sewage disposal plant in Lanzhou Petrochemical Company, PetroChina in Gansu Province, China (36°10′N, 103°67′E). The tank treats 38,200 m^3 of refinery wastewater per day using A/O + O (anaerobic/aerobic + aerobic) processes. Dissolved Oxygen (DO), pH, and sampling temperature were recorded *in situ* with a parameter recorder. After sampling, the activated sludge and wastewater were kept in sterilized boxes/carboys and then transported to the laboratory immediately for further experiments. In the lab, 1 L activated sludge and 10 L wastewater were used for characteristic analyses. The concentrations of petroleum were detected using gas chromatography-flame ionization detector (GC-FID) (Wang et al., 1994; Wang et al., 1995). Other parameters such as COD, 5-day biochemical oxygen demand (BOD_5), methanol concentration, volatile phenol, ammonical nitrogen, cyanide, sulphide, suspended solids (SS) and turbidity of water were measured as per the methods described previously (Huang et al., 2014; Ródenas-Torralba et al., 2007). Concentrations of chromium (Cr), nickel (Ni), zinc (Zn), cadmium (Cd), lead (Pb), arsenic (As) and mercury (Hg) were determined by atomic absorption spectrometry technique (Olmedo et al., 2010). The settled volume (SV_{30}), sludge volume index (SVI) and mixed liquor suspensions of activated sludge (MLSS) were measured according to the standard methods (Zhao et al., 2014). Each parameter was detected in triplicate (Table S1).

The remaining activated sludge was used for immobilization using a continuous aeration method. In this study, nano-attapulgite clay compounded hydrophilic urethane foam (AT/HUFs), which is a kind of foamy nanomaterial, was used for the immobilized supports (Zhang et al., n.d.). During immobilization, the hydraulic retention time (HRT) was 24 h. Each time the COD concentration was recorded (daily), 10% of the wastewater was changed. After 2 weeks of incubation, the removal rate of COD reached 30%, which indicated that the immobilization was completed.

2.2. Operation of BF-MBR system

The operation was carried out in a continuous BF-MBR with a working volume of 25 L (Fig. 1). The three tanks of the BF-MBR included a raw-wastewater tank, reaction tank and sterilization tank which were