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Process Safety and Environmental Protection



journal homepage: www.elsevier.com/locate/psep

# The effects of different carriers on removal performance and membrane fouling by HMBR in treating sewage with low carbon-to-nitrogen ratio



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#### ARTICLE INFO

Article history: Received 30 November 2015 Received in revised form 3 June 2016 Accepted 11 June 2016 Available online 17 June 2016

Keywords: Carrier HMBR Sewage Membrane fouling Removal performance EPS

# ABSTRACT

A hybrid membrane bioreactor (HMBR) with biological-band carriers (reactor A) and a HMBR with suspended-honeycomb carriers (reactor B) were used for treatment of sewage with low carbon-to-nitrogen ratio (3–5). The pollutant removal performance and membrane fouling in HMBRs was investigated under different hydraulic retention times (HRT) (5 h and 9 h). The results demonstrated that both HMBRs can be effective for the removal of chemical oxygen demand (COD), NH<sub>3</sub>-N and total nitrogen (TN). The rate of TN removal in reactor A was higher than that in B. Moreover, it was found reactor A was more advantageous for the degradation of biodegradable organic matter than that of B. The biomass in reactors was characterized by mixed liquor adherent solids (MLAS), particle size distributions, mixed liquor suspended solids (MLSS) and concentration of extracellular polymeric substance (EPS). The results showed that MLAS content and particle size distribution in reactor A were higher than in B, whereas concentrations of MLSS and EPS exhibited the opposite trend. When systems were run under different HRTs, the increased rate of trans-membrane pressure (TMP) in reactor A was slower than that in B. Therefore, reactor A was considered superior to reactor B in pollutant removal and controlling membrane fouling.

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## 1. Introduction

Membrane bioreactor (MBR), an innovative and promising option for wastewater treatment, has drawn special attention since 1980s (Kin and Jang, 2006; Wang et al., 2006; Miura et al., 2007). However, its widespread application was restricted because of two main concerns related to membrane fouling, and low nitrogen removal efficiency. During the last few years, many researchers have discovered that a novel hybrid membrane bioreactor (HMBR) with biological carriers in MBR can reduce the sludge yield to some extent, intensify the denitrification effect and control membrane fouling (Cao et al., 2004). Paetkau and Cicek (2011) found that the TN removal rate was greater than 80% in HMBR while 31% in MBR during treatment synthetic wastewater, suggesting that suspended and immobilized microbes in HMBR can significantly enhance the synchronous nitrification and denitrification process. In addition, it was also found that HMBR can relieve membrane pollution by improving the activated sludge properties and microbial activity. Hu et al. (2012) found that the concentration of EPS was 76% lower in HMBR installed with a biological carrier as opposed to that in MBR, which immensely alleviated membrane fouling.

There are various types of biological carriers that can be installed in a HMBR system. According to the mode of operation, a biological carrier can be divided into two broad categories, suspended carriers, and fixed installed carriers. Suspended carriers have fluidized movement with gas-liquid two-phase flow in a reactor, including the microorganisms on the carrier that also move. During the operational process, suspended carriers were scrubbed with a gas-liquid two-phase flow, and their microbes collided and rubbed each other, which led to the limited thickness of adhesion biofilms (Seifi, 2012; Xia et al., 2008). Fixed installed carriers were generally composed of flexible materials. The

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http://dx.doi.org/10.1016/j.psep.2016.06.019

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carrier was fixed in the biochemical reaction basin with one fixed end while the other end moved with a gas-liquid two-phase flow. This type of installation reduces the shear force on the carrier, resulting in the greater thickness of the adhesion biofilm and the rich biological phase (Feng et al., 2008; Nguyen et al., 2012). In terms of controlling membrane fouling and nitrogen removal efficiency, most studies indicate that the two modes of HMBR are more advantageous than traditional MBR (Yang and Yang, 2011; Lim et al., 2011). Little is known about the differences between the two modes of HMBR. However, a suspended carrier or a fixed installed carrier has its own characteristics in a HMBR. The two modes are reported to be different due to certain factors that affect their performance, such as the state of microorganism, MLSS, MLAS and particle size distribution in a HMBR reactor (Yang et al., 2009), and hence, require a further in-depth study.

In recent years, there is a trend of low carbon-to-nitrogen ratio in municipal domestic wastewater in China, which results in serious problems related to the denitrification of wastewater. The carbonto-nitrogen ratio is a key parameter in biological nitrogen removal, which directly affects competition between autotrophic bacteria and heterotrophic bacteria (Michaud et al., 2006). The reason for the undesirable TN removal efficiency using a traditional two-stage process is a lack of the carbon source, which inhibits the growth of denitrifying bacteria. MBR was also applied in treating sewage with low C/N ratio, but its denitrification performance is poor. As a completely mixed aeration basin, it is difficult to guarantee the stability of the micro anaerobic environment in MBR, so the growth of denitrifying bacteria is also restrained similarly. HMBR combines the contact oxidation method and activated sludge processes. The thicker biological membrane can maintain a micro anoxia or anaerobic environment when dissolved oxygen (DO) is higher in reactor, which is conducive to the growth of denitrifying bacteria, thereby improving the TN removal efficiency.

In this study, two HMBR reactors with different carriers were used for the treatment of sewage with low carbon-to-nitrogen ratio (3–5). Based on the results of our team's previous operation, the TN and COD removal efficiencies in both HMBRs under different hydraulic retention times (HRT) (5 h and 9 h) were investigated. The ability of the two HMBRs to degrade dissolved organic matter (DOM) was also estimated using a three-dimensional fluorescence spectrum-area integral method. The microscopic properties of mixed liquor in both HMBRs were analyzed, such as MLAS, particle size distributions, MLSS and concentration of EPS. Meanwhile, possible sources of membrane fouling were investigated throughout the operation for both types of HMBRs.

# 2. Materials and methods

## 2.1. Experimental setup and operation parameter

Two identical 77 L (the carrier area was 28.7 L, and the membrane module area was 38.5 L) HMBR systems (Fig. 1) were run in parallel during the study. In reactor A biological band with 15% packing ratio was added in its carrier area, and in reactor B suspended honeycomb carriers with 15% packing ratio was added. A polypropylene hollow fiber membrane unit with a pore size of 0.1  $\mu m$  and a filtration area of 0.2  $m^2$  was immersed in each HMBR membrane module area, with an operating pressure of 10–40 kPa and a membrane flux of 20 L/m<sup>2</sup> h. The aeration system was located at the bottom of the carrier area and the membrane module area in each HMBR. The influent was supplied continuously through U-tube water level box. The effluent was intermittent with a cyclic pumping mode (10 min on and 3 min off). This 60 days experiment was run in two stages with no sludge discharge during the first 30 days, stage 1: HRT for each reactor was set for 9 h, while during the next 30 days, stage 2: HRT was set for 5 h. The new membrane module was replaced on the 30th day of the experiment. DO concentration was maintained at 4.0 mg/L throughout the procedure. To stimulate the growth of biofilm, mixed strain was

applied. Once the biofilm growth was stabilized, both HMBRs were seeded with activated sludge from sewage treatment plant.

#### 2.2. Characteristics of carriers and raw wastewater

In reactor A, a flexible biological band was used as the carrier, while in reactor B, suspended honeycomb was used as the carrier. The physical parameters of these carriers are shown in Table 1. Raw wastewater was collected from a school sewage outfall, and its characteristics are shown in Table 2.

# 2.3. Analytical methods

#### 2.3.1. Measurement of conventional indicators

NH<sub>3</sub>-N, TN, COD, MLSS, MLAS, pH and DO were measured by using their respective laboratory standard methods. NH<sub>3</sub>-N was measured according to standard method using visible spectrophotometer (V-5600, Metash Inc., China) (Chinese HJ535-2009). Total nitrogen (TN) was determined using an ultraviolet-visible spectrophotometer (UV-2450, Shimadzu Inc., Japan) (Chinese GB11894-89). COD was measured using the potassium dichromate method (Chinese GB11914-89). pH values, DO and water temperature were measured by using a Dissolved Oxygen Analyzer (YSI550A, YSI Inc., USA). MLAS should be stripped from the carrier by ultrasonic oscillations. MLSS and MLAS were measured in accordance with the standard method (gravimetric method). To estimate the DOM content, three-dimensional fluorescence spectrum-area integral method was used. Suspended sludge particle size was measured using a Laser Particle Analyzer (S3500, Microtrac Inc., USA).

#### 2.3.2. Extraction and determination of EPS

For the extraction of EPS, mixed liquor samples were collected from the aeration room of both HMBRs when the systems were stable. The extraction of EPS was carried out in accordance with the standard centrifugation procedure as reported by Arabi and Nakhla (2009), with minor modification. In short, the mixed liquor samples were initially centrifuged for 20 min at 12,000  $\times$  g (4 °C). The supernatant obtained from each sample was filtrated through 0.45  $\mu m$  cellulose acetate membrane and a 3500 Da membrane. The concentration of the extracted EPS of each sample was measured by using three-dimensional fluorescence spectroscopy (3DEEM). The area integral method was then adopted to process the data. A literature survey revealed that the contents of IV and V in the fluorescence spectra might be the representatives of the concentration of EPS (Guo et al., 2014). All 3DEEM spectra were measured with a luminescence spectrometer (LS-55, Perkin-Elmer, Japan). In this study, the EEM spectra were collected with corresponding scanning emission spectra from 280 nm to 550 nm at 0.5 nm increments by varying the excitation wavelength from 220 nm to 400 nm at 10 nm sampling intervals. The excitation and emission slits were maintained at 10 nm and the scanning speed was set at 1200 nm/min in this study. The spectra of Milli-Q water was recorded to eliminate water Raman scattering and to reduce other background noise. A 290 nm emission cutoff filter was used in scanning to eliminate the second order Raleigh light scattering.

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