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Discrepant membrane fouling of partial nitrification and anammox membrane bioreactor operated at the same nitrogen loading rate

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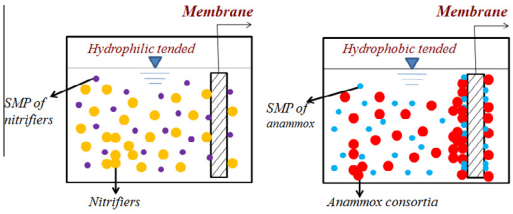
HIGHLIGHTS

- Anammox–MBR has a more serious membrane fouling than PN–MBR.
- It caused by different microbial products of nitrifiers and anammox bacteria.
- Anammox bacteria metabolites are more hydrophobic than that of nitrifiers.
- Hydrophobic PVDF membrane absorbs more hydrophobic anammox bacteria than nitrifiers.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, two times more serious membrane fouling was found in anammox membrane bioreactor, compared to partial nitrification membrane bioreactor (PN–MBR) operated at the same nitrogen loading rate. By protein, polysaccharide, amino acids and functional groups analysis, it was found that the discrepancy in membrane fouling was virtually due to the difference in microbial products of nitrifiers and anammox bacteria. Protein and polysaccharide were main foulants on membrane surface; meanwhile theirs content and ratio in the EPS, supernatant and membrane surface were significantly different in PN–MBR and anammox–MBR. The anammox metabolism products contained much more hydrophobic organics, hydrophobic amino acids, and hydrophobic functional groups than nitrifiers. A mass of anammox bacteria as well as hydrophobic metabolism products deposited on the hydrophobic membrane surface and formed serious fouling. In further, hydrophilic modification is more urgently needed to mitigate membrane fouling when running anammox–MBR, than PN–MBR.

1. Introduction

The partial nitrification (PN)–anaerobic ammonium oxidation (anammox) process is regarded as a new, efficient solution to remove ammonium from wastewater without organic carbon consumption (Liang et al., 2014). In this process, ammonium oxidizing bacteria (AOB) oxidize half of the ammonium to nitrite, and then the remained ammonium and nitrite are converted to nitrogen gas by anammox bacteria (Van de Graaf et al., 1996). PN–anammox process is a novel nitrogen removal process with inherent superiority of no need external carbon source, less required oxygen and reduced sludge production etc, compared to the conventional nitrification and denitrification process (Jetten et al., 2002).

The functional bacteria, AOB and anammox bacteria both have a low proliferation rate (Van de Graaf et al., 1996), which results in a long term period to start up reactors. Thus, the optimization of

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reactor configuration and operation conditions is essential. Recently, membrane bioreactors (MBRs) have shown great potential in running the PN–anammox process (Shen et al., 2014). MBR allows a complete separation of hydraulic retention time (HRT) and sludge retention time (SRT) by the use of membrane filtration (Zhang et al., 2015), so it could achieve high biomass concentration and establish a proper environment for the cultivation of these bacteria (Xue et al., 2009).

Although MBR has lots of advantages, membrane fouling is a non-ignorable problem during its operation. Therefore, the membrane fouling of PN–MBR and anammox–MBR has become a current research focus (Zhang et al., 2009; Shen et al., 2014). For PN–MBR and anammox–MBR, it has been found that reactor operation period plays a crucial role in membrane fouling. They have very low transmembrane pressure (TMP) values at the beginning period of reactor operation owing to the low sludge concentration. With time course of reactor operation, the increase of soluble microbial products (SMP) lead to the elevated fouling rates; meanwhile, the ratio of polysaccharide and protein in EPS of membrane surface increased following a more serious membrane fouling. The crosslinked structure of polysaccharide formed a gel layer on membrane surface more easily than protein to cause membrane fouling (Shen et al., 2014).

Actually, different types of microorganisms own variation of metabolites, finally resulted in different extracellular polymeric substances (EPS) and SMP (Gao et al., 2014), which can be derived in the excreting process of bacteria during substrate metabolisms, biomass growth, and biomass decay (Jarusutthirak and Amy, 2006). As reported by Hou et al. (2015), the microbial products by AOB and anammox are clearly different. Anammox bacteria are more hydrophobic than activated sludge, nitrifying bacteria and denitrifying bacteria, and contain more hydrophobic functional groups in extracellular polymers (Hou et al., 2015). Many reports have claimed that EPS and SMP mainly composed of proteins and polysaccharides is the key contributor to the high membrane fouling in MBR (Nguyen et al., 2014; Luo et al., 2015). EPS has a high correlation with composition change of a cake layer, which significantly affects membrane fouling conditions (Nguyen et al., 2014). Thus, different types of microorganisms are supposed to cause different membrane fouling performances.

Accordingly, there is a question raised that whether the difference in characteristics of microbial products between nitrifiers and anammox bacteria affects membrane fouling. Therein, the purpose of this study was to compare membrane fouling properties of PN–MBR and anammox–MBR operated at the same nitrogen loading rate and analyze the reasons for different fouling levels.

In this study, PN–MBR and anammox–MBR were operated at the same nitrogen loading rate. Significant discrepancies were verified in the aspect of membrane fouling levels. To go inside for the mechanism, excitation–emission matrices (EEM), protein and polysaccharide analyses for supernatant were explored to validate the difference in microbial products in the two reactors. Proteins, polysaccharides, amino acids and organics functional groups on membrane surface analyses were further explored to clarify the hydrophobic property of organic matter produced by nitrifiers and anammox bacteria and the different production of these metabolisms. Most importantly, a conclusion was drawn that the different metabolites produced by these two consortia significantly contributed the discrepant membrane fouling profile.

2. Materials and methods

2.1. Reactor design and operation

PN–MBR and anammox–MBR have the same size and cylindrical shape with an effective volume of 5.0 L (Fig. 1). The abbreviations for the nomenclatures in this study have been shown in Table 1. The membrane was made of polyvinylidene fluoride (PVDF) with an effective surface area of 0.11 m^2 (Yue Qing Membrane Technology Co., Ltd, China). The hollow fiber membrane with a pore size of 0.03 μm was arranged in the center of the reactor. No modification was applied on for the PVDF membrane surface. The contact angle of this PVDF membrane and water was 79.4 ± 1.0° identifying by contact angle analyzer (Dataphysics Co., Germany), similar to the reference reported (Ochoa, 2003; Yan et al., 2006). A baffle between the main body of reactor and the membrane surface was arranged in these reactors to avoid the direct contact of aeration device and membrane surface. Anammox–MBR was inoculated with anammox bacteria (Liu et al., 2015), and PN–MBR was inoculated with nitrifiers sampled from an aerobic bioreactor.

These two reactors were fed with a synthetic medium. Solutions containing $(NH_4)_2SO_4$, $NaNO_2$ and some trace elements were added to the anammox–MBR. The trace element composition has been described previously (Van de Graaf et al., 1996). PN–MBR had the same influent composition except that all nitrogen was introduced in the form of $(NH_4)_2SO_4$. Besides the initial few days for the start-up, the nitrogen loading rate was controlled at 300 mg N/L day for both of the reactors, which were run for 50 days with HRT of 24 h. The temperature and pH of both reactors were controlled at 7.4–8.3 and 37 ± 0.5 °C. The anaerobic (Dissolved Oxygen <0.2 μmol/L) and micro aerobic environments (Dissolved Oxygen 1 mg/L) were maintained correspond to anammox–MBR and PN–MBR. The stirring speed of both reactors was set at approximately 100 rpm. The effluent was filtered by the membrane module driven by a peristaltic pump.

2.2. General water quality parameters

Samples were taken every two days to monitor the influent and effluent quality. The concentrations of ammonium, nitrite and nitrate were measured according to standard colorimetric methods, as set out by the American Publish Health Association (APHA, 1995). The pH was determined with a pH meter (TP–110, Mettler, China). The Dissolved oxygen (DO) level was measured by a DO meter (DOS–328B Mettler, China). TMP was measured by a Vacuum Pressure Gauge (MD–S600, MEOKON, China) with a range of from −0.1 MPa to 0.1 MPa.

The reactor's mixture liquid (10 mL) was withdrawn from the reactor once a week and centrifuged for 10 min at 3000 rpm to obtain the supernatant. Dissolved organic matter (DOM) was obtained by filtering the supernatant by a cellulose acetate membrane filter with a pore size of 0.45 μm (Tianjin Dongtang Technology Co., Ltd, China). EPS were extracted based on cation ion exchange resin method (Oslash et al., 1996). The protein and polysaccharide in supernatant, DOM and EPS could be determined by a phenol–sulfuric acid method (Dubois et al., 1956) and Lowry method (Lowry et al., 1951), respectively, and all the analyses were performed for three times to get the average value. In addition, EEM analysis was applied to depict the organic compounds in the supernatant. EEM spectrograph (F7000, Hitachi Limited, Japan) was applied here. The EEM spectra were collected by scanning emission wavelengths from 300 nm to 700 nm. The excitation wavelength was increased from 200 nm to 600 nm at 10 nm increments. The scanning speed was set at 1200 nm/min.

2.3. Membrane surface analysis

All the samples for membrane surface analysis were probed at the terminal operation day (day 50) of PN–MBR and anammox–MBR.