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Efficient methanogenic degradation of alcohol ethoxylates and microbial community acclimation in treatment of municipal wastewater using a submerged anaerobic membrane bioreactor

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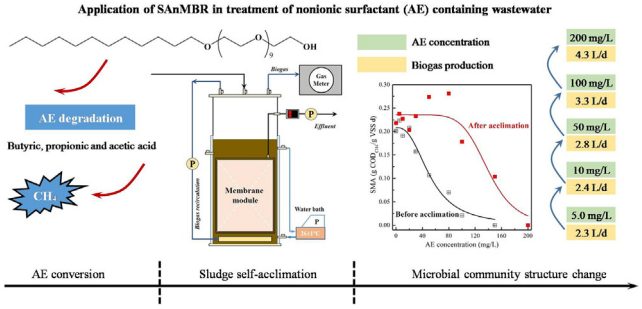
HIGHLIGHTS

- AE was all degraded and converted into CH_4 by anaerobic microbes in SAnMBR.
- Sludge acclimation to AE greatly changed the microbial community structure.
- Tolerance ability of methanogenic activity to AE was greatly enhanced.
- Higher concentration of AE caused an increase of membrane fouling rate.

GRAPHICAL ABSTRACT



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ABSTRACT

The effect of alcohol ethoxylates on the treatment of municipal wastewater by a submerged anaerobic membrane bioreactor was investigated by a 400 days operation including the treatment efficiency, methanogenic activity of sludge and microbial community structure. The results indicated that alcohol ethoxylates (5.0–200 mg/L) was efficiently degraded and converted into methane due to the similar COD removal 95.5–98.8% and rising biogas production rate (2.30–4.25 L/d) compared with control (96.8% and 2.55 L/d). The microbes in sludge could copy with the presence of alcohol ethoxylates in wastewater by releasing more SMP and EPS, which caused a higher membrane fouling rate. Moreover, via long term acclimation, the specific methanogenic activity of sludge was greatly enhanced due to the changes of microbial community structure. Hence, the sludge self-acclimation to alcohol ethoxylates was responsible to the efficient methane recovery in treatment of municipal wastewater.

1. Introduction

Alcohol ethoxylates (AE) has been used as an ideal replacement of alkylphenol ethoxylates (APE) due to the potential estrogenicity of APE degradation products (Merrettig-Bruns and Jelen, 2009), it is hence used widely in domestic and commercial detergents, household and personal care products (Traverso-Soto et al., 2013). However, the presence of AE in aquatic environment could cause acute and chronic effects in sensitive organisms, such as crustacean and

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fish (Ferraraa et al., 2005). Although AE can be efficiently removed by physical and chemical processes, such as adsorption and AOPs (Potapova et al., 2014; Da Silva et al., 2015), their cost is very high and chemical oxidation just destroys its initial molecule but generates carboxylic by-products. While the conventional processes for efficient treatment of AE containing wastewater, such as aerobic and anoxic techniques, are also not sustainable due to their energy intensive, large qualities of residuals production and failure to recover the potential resources available in wastewater (Martinez-Sosa et al., 2011; Gao et al., 2014).

The anaerobic process is currently recognized as a promising technology due to its lower energy consumption, low sludge production and biogas generation (Hahn and Figueroa, 2015). Among the different anaerobic processes, the anaerobic membrane bioreactor (AnMBR), by coupling membrane filtration with anaerobic treatment, provides an alternative strategy for wastewater treatment with nearly absolute biomass retention and the potential to generate a higher quality effluent (Ng et al., 2015). At present, AnMBR has been successfully studied for the treatment of high strength wastewater, such as food and beverage industry wastewater and swine manure (Ng et al., 2015). Only recently, it has been used to treat municipal wastewater (Smith et al., 2012; Ozgun et al., 2013) and the influence of temperature, HRT, OLR, membrane characteristics on reactor performance has also been investigated in detail (Ozgun et al., 2013).

Although the degradation of high concentration of alkylphenol ethoxylates (APE) and its toxicity or inhibition effect on the anaerobic microorganism have been studied (Song and Bielefeldt, 2012; Bozkurt and Sanin, 2014), however, the influence of alcohol ethoxylates (AE) on the microbe is little known since the increased consumption and production of AE compared with APE (Motteran et al., 2014a,b). Besides, an anaerobic fluidized bed reactor rather than AnMBR was used in the above studies. AE is considered biodegradable, while high rates of instabilities in biological process may occur in anaerobic treatment of wastewater containing these surfactants. Hence, it is also necessary to investigate the AE effect on AnMBR performance and methane production potential of sludge during municipal wastewater treatment. At present, the most widely accepted degradation pathway of AE under anaerobic condition involves the central fission of the ether bond between the alkyl and ethoxylated chain, resulting in the formation of fatty acids and PEGs (Huber et al., 2000). However, the conversion of PEG into smaller organic acids and then to methane is still lack of direct evidence. In general, few studies were conducted to explore the suitability of AnMBR in treatment of municipal wastewater if AE was present including the treatment efficiency, methanogenic activity of sludge and the response of microbial community structure. It is very important for the application of AnMBR in practical wastewater treatment.

In this study, the influence of AE on AnMBR performance during municipal wastewater treatment was firstly investigated via long-term operation, such as the COD removal, biogas production, SMP/EPS generation, membrane fouling and sludge concentration. On the other side, sludge self-acclimation to alcohol ethoxylates on the specific methanogenic activity was further evaluated by comparing their difference before and after long term acclimation. Hence, the microbial diversity distribution may changed after long term acclimation in treatment of alcohol ethoxylates containing wastewater, which led to higher methane recovery efficiency. Finally, the role of AE on the anaerobic microbes and the response of microbial community structure to AE presence were investigated and discussed by the pyrosequencing method. The findings in this study are expected to provide some useful information that whether SAnMBR is suitable to dispose the AE containing municipal wastewater.

2. Experimental and methods

2.1. SAnMBR setup and operation

As shown in Fig. S1, a submerged anaerobic membrane bioreactor (SAnMBR) with a working volume of 6 L was operated at 25 ± 1 °C using a flat-sheet submerged membrane (Kubota Corporation, Osaka, Japan). The membrane that fixed in the lower part of SAnMBR was made of chlorinated polyethylene with a non-woven fibrous support (polyethylene terephthalate: PET), which has a normal pore size of 0.2 μm and a total area of 0.116 m^2. A coarse tube diffuser was located below the membrane. Biogas in the headspace was recirculated by a pump at a flow rate of 5 L/min to provide membrane hydrodynamic shearing and reactor mixing. Permeate was suctioned by a peristaltic pump. transmembrane pressure (TMP) was measured by a pressure sensor located on the permeate line. The reactor was warmed by water circulation. A wet gas meter was used to measure the amount of daily biogas. The characteristics and composition of the synthetic municipal wastewater were shown in Table S1. The substrate tank was stirred (200–300 rpm) to keep the substrate at a uniform state. The reactor was inoculated with waste activated sludge from the municipal sewage treatment plant (Sendai, Japan).

The long-term experiment was divided into stage I (without AE, 80 days, 2 phases), stage II (with AE 5 and 10 mg/L, 247 days, 3 phases) and stage III (with AE 50–200 mg/L, 64 days, 3 phases). The reactor operation at different phases were summarize in Table 2. Fig. S2 showed the structure of used AE (decaethylene glycol mono-dodecyl ether, Sigma-Aldrich). Membrane was changed once the TMP reached above 30 kPa. After putting back the cleaned membrane, air in the reactor headspace was replaced by nitrogen gas.

2.2. Specific methanogenic activity of sludge and batch experiment

The specific methanogenic activity (SMA) of sludge and batch experiments were all carried out in 120 mL serum bottles using acetate, H_2/CO_2 and different amount of AE. In the SMA test, after the addition of 40 mL sludge and 40 mL nutrient solution containing trace element (as shown in Table 3), the serum bottles were sealed with rubber stoppers and secured by aluminum crimp. Oxygen in headspace of the bottles was purged with nitrogen gas for 2 min. Prior to use, the nutrient solution was boiled for 2 h to remove any dissolved oxygen present and cool down to room temperature under nitrogen atmosphere. The initial COD concentration for the vials added with acetate was all 2000 mg/L. For H_2/CO_2, the headspace of the bottle was replaced with pressurized gas of H_2/CO_2 (80:20, v/v) to get a final pressure of 1.4 atm. Then, 1 mL $Na_2S{\cdot}9H_2O$ (250 mg/L as a final concentration in the vial), used as the reducing agent, was injected into each bottle to obtain an absolutely anaerobic condition. Finally, different amount of AE stock solution was added into the above bottles, which were placed and incubated in a water bath (100 ± 1 rpm) at 25 ± 1 °C. After each of the bottles reached the set temperature, the headspace was vented using a syringe to release the pressure caused by the thermal expansion. Biogas production and composition were measured every 3–5 h according to the biogas volume, and expressed as the value at the standard state. Each experiment was conducted in two replicates to ensure its reliability.

AE degradation kinetics was further investigated in the presence or absence of acetate, respectively. Biogas production and composition were measured every 4–6 h. At the same time, 2.5 mL of sample was withdrawn and centrifuged to remove sludge and AE concentration was then monitored. The other experimental procedure was the same as described above.