



# Effects of dissolved organic matters (DOMs) on membrane fouling in anaerobic ceramic membrane bioreactors (AnCMBRs) treating domestic wastewater



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## ARTICLE INFO

### Article history:

Received 7 January 2015

Received in revised form

6 July 2015

Accepted 21 July 2015

Available online 23 July 2015

### Keywords:

Anaerobic membrane bioreactor

Ceramic membranes

Dissolved organic matters

Membrane fouling

Low-strength wastewater

Pore size

## ABSTRACT

Anaerobic membrane bioreactors (AnMBRs) have been regarded as a potential solution to achieve energy neutrality in the future wastewater treatment plants. Coupling ceramic membranes into AnMBRs offers great potential as ceramic membranes are resistant to corrosive chemicals such as cleaning reagents and harsh environmental conditions such as high temperature. In this study, ceramic membranes with pore sizes of 80, 200 and 300 nm were individually mounted in three anaerobic ceramic membrane bioreactors (AnCMBRs) treating real domestic wastewater to examine the treatment efficiencies and to elucidate the effects of dissolved organic matters (DOMs) on fouling behaviours. The average overall chemical oxygen demands (COD) removal efficiencies could reach around 86–88%. Although CH<sub>4</sub> productions were around 0.3 L/g COD<sub>utilised</sub>, about 67% of CH<sub>4</sub> generated was dissolved in the liquid phase and lost in the permeate. When filtering mixed liquor of similar properties, smaller pore-sized membranes fouled slower in long-term operations due to lower occurrence of pore blockages. However, total organic removal efficiencies could not explain the fouling behaviours. Liquid chromatography-organic carbon detection, fluorescence spectrophotometer and high performance liquid chromatography coupled with fluorescence and ultra-violet detectors were used to analyse the DOMs in detail. The major foulants were identified to be biopolymers that were produced in microbial activities. One of the main components of biopolymers – proteins – led to different fouling behaviours. It is postulated that the proteins could pass through porous cake layers to create pore blockages in membranes. Hence, concentrations of the DOMs in the soluble fraction of mixed liquor (SML) could not predict membrane fouling because different components in the DOMs might have different interactions with membranes.

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## 1. Introduction

Anaerobic processes have been widely regarded as a possible way to achieve economic sustainability and energy neutrality in wastewater treatment plants (Liao et al., 2006). Much less waste biomass is produced compared to aerobic processes due to the slow growth rates of anaerobic microorganisms (Stuckey, 2012). Nonetheless, their slow growth rates could create challenges in treating wastewater, especially in start-up periods, due to washout of these slow growing microorganisms (Van Haandal and Lettinga, 1994). Anaerobic membrane bioreactors (AnMBRs) that couple

membranes into the anaerobic processes not only fully retain the anaerobic microorganisms but also produce an effluent with high quality (Stuckey, 2012). As solids-liquid separation is perfect and sedimentation can be eliminated, footprints of the treatment plants are greatly reduced (Kanai et al., 2010). In spite of being an attractive option for wastewater treatment, AnMBRs have not been studied extensively with real domestic wastewater. The biggest hindrances in adopting AnMBRs in domestic wastewater treatment include (1) membrane fouling in AnMBRs is not well understood; and (2) the energy produced from low organic contents may not be able to heat up the anaerobic reactors, especially in cold climate (McCarty et al., 2011). This problem can be exacerbated by dissolved CH<sub>4</sub> in the permeate.

Polymeric membranes are the most widely used membranes in membrane bioreactors (MBRs), including AnMBRs, due to their low

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## Nomenclature

AeCMBR	aerobic membrane bioreactor	MBR	membrane bioreactor
AnCMBR	anaerobic ceramic membrane bioreactor	MLSS	mixed liquor suspended solids
AeMBR	aerobic membrane bioreactor	MLVSS	mixed liquor volatile suspended solids
AnMBR	anaerobic membrane bioreactor	MW	molecular weight
BW	backflushing water	MWD	molecular weight distribution
COD	chemical oxygen demand	PSD	particle size distribution
DOM	dissolved organic matter	R <sub>s</sub>	COD removal efficiency in mixed liquor supernatant
FL	fluorescence response	R <sub>t</sub>	COD removal efficiency in membrane permeate
HPO	hydrophilic organics	SEM	scanning electronic microscopy
HPI	hydrophilic organics	SML	soluble fraction of mixed liquor
HPLC	high performance liquid chromatography	SRT	solids retention time
HRT	hydraulic retention time	tDOC	total dissolved organic carbon
LC-OCD	liquid chromatography-organic carbon detection	TN	total nitrogen
LMWs	low molecular weight substances	TOC	total organic carbon
		UV <sub>220</sub>	UV absorbance at wavelength of 220 nm

manufacturing cost (Skouteris et al., 2012) and availability of operational experience, particularly for aerobic MBRs (AeMBRs). However, new research fields have increasing demands for chemically and thermally stable membranes (Caro et al., 2000). Thus, inorganic membranes such as ceramic membranes have attracted increasing attention in recent years despite their high upfront capital costs. Lee et al. (2013) found that ceramic membranes have lower fouling propensity than polymeric membranes due to the weaker bonding between foulants and the membranes. In addition to their low fouling propensity, more aggressive cleaning agents can be used to shorten cleaning time required due to their excellent stability and integrity. As a result, the physical and chemical cleaning efficiencies could be increased by 75% and 25%, respectively (Lee and Kim, 2014). Other than these advantages, laborious maintenances such as repair and replacement of polymeric hollow fibres could be eliminated (Lee et al., 2013). All these above-mentioned advantages of ceramic membranes suggest that anaerobic ceramic membrane bioreactors (AnCMBRs) may have great potential for wide-scale application if the cost of ceramic membranes can be reduced and when life-cycle cost is being considered.

Most of the ceramic membranes used in AnMBRs are tubular-shaped owing to their low fouling propensity that was resulted from shear produced by cross flow (Lin et al., 2013). However, as these systems were operated under pressure-driven mode (Herrera-Robledo et al., 2009), there might be a risk of more severe fouling due to shearing of microorganisms by high pressure pumps (Choo and Lee, 1996). This can create greater issues in AnMBRs than in AeMBRs as the slow-growing anaerobic microorganisms may not be able to recover fast enough to compensate for the loss. Moreover, the energy consumption for pressure-driven mode is higher than that for the submerged mode due to higher pressure required (Liao et al., 2006). In this sense, submerged configuration that relies on vacuum suction may be more favourable for AnMBRs.

Despite the great potentials of AnMBRs and ceramic membranes, membrane fouling remains as the biggest challenge as it increases operating and capital costs (Martinez-Sosa et al., 2011). Therefore, understanding membrane fouling in AnMBRs is essential in order to find proper controlling methods. In membrane fouling studies, several parameters including operating conditions, feedwater-biomass characteristics and membrane characteristics are considered to have major impacts on membrane fouling (LeClech et al., 2006). Studies on the feedwater-biomass characteristics usually attributed fouling to mixed liquor suspended solids (MLSS) and soluble fraction of mixed liquor (SML). In AeMBRs, conflicting opinions on the effects of MLSS on membrane fouling

have been reported (Brookes et al., 2006; Chang and Kim, 2005; Rosenberger et al., 2006). Likewise, there is no consensus on the impacts of SML on membrane fouling. Several authors reported that SML had negative impacts on the membrane fouling (Bouhabila et al., 2001; Rosenberger et al., 2006), while others could not find this correlation (Drews et al., 2008; Kimura et al., 2009). The disparity might arrive from the fact that some components in SML had larger impacts on membrane fouling than the others (Miyoshi et al., 2012). These two factors were studied to a much lesser extent in AnMBRs. Nonetheless, similar results on the effect of MLSS concentration could be found in available literatures of AnMBRs (Lin et al., 2010; Robles et al., 2013). Most of the studies that investigated the SML in AnMBRs used synthetic feedwater to facilitate the examination of the soluble microbial products (Stuckey, 2012). However, influent organics could not be ignored when real wastewater was used as membrane fouling rates were reported to be closely related to the influent chemical organic demand (COD) levels (Lin et al., 2010).

To date, few studies have been reported on the treatment of real domestic wastewater using ceramic membranes in AnMBRs. Species of the dissolved organic matters (DOMs) in the SMLs were not fully understood, while their effects on membrane fouling were seldom reported in AnMBRs. In this study, AnCMBRs were fed with real domestic wastewater to investigate the effect of membrane pore size on DOMs production and its subsequently effects on membrane fouling. While most the studies for fouling mechanisms relied on statistical correlations between fouling rates and concentrations of foulants in SML, foulants were recovered from the cake layers and the pores, and subsequently, characterised in this study.

## 2. Materials and methods

### 2.1. Reactors setup

Three acrylic AnCMBRs, each with an effective volume of 3.6 L, were set up in parallel (Fig. 1). They were denoted as R80, R200 and R300 for AnCMBRs, in which 80, 200 and 300 nm pore-sized ceramic (Al<sub>2</sub>O<sub>3</sub>) membranes were immersed in, respectively. They were operated under ambient temperature of 25–30 °C, which was in the mesophilic range. The HRT and SRT were maintained at 7.5 h and 60 d, respectively, in all the three AnCMBRs. The biogas produced was collected by water displacement method and was recirculated to scour the membrane surface through two gas diffusers by a KNF compressor (KNF, N86 KT 18, Germany) in each

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