Bioresource Technology 196 (2015) 586–591

Contents lists available at ScienceDirect

Bioresource Technology

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

Structure and distribution of inorganic components in the cake layer of a membrane bioreactor treating municipal wastewater

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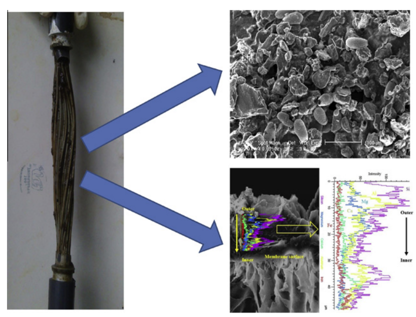
HIGHLIGHTS

- Characterize the structure and distribution of the inorganic in cake layer of MBR.
- BCR, XPS and map/line scanning EDX identify the inorganic structure in cake layer.
- Si, Al, Ca, Mg, Fe, and Ba are the predominant inorganic compounds in cake layer.
- The predominant inorganic compounds are found to occur mostly as crystal particles.
- Si and Al accumulate together along the cross-sectional cake layer as SiO_2–Al_2O_3.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 10 June 2015
Received in revised form 3 August 2015
Accepted 8 August 2015
Available online 14 August 2015

Keywords:
Membrane bioreactor
Cake layer
Inorganic distribution
BCR sequential extraction
SiO_2–Al_2O_3 crystal particles

ABSTRACT

A laboratory-scale submerged anoxic–oxic membrane bioreactor treating municipal wastewater was operated to investigate the structure and distribution of the inorganic cake layer buildup on the membrane. BCR (European Community Bureau of Reference) sequential extraction, X-ray photoelectron spectroscopy (XPS), and both map and line scan of energy-dispersive X-ray analysis (EDX) were performed for cake layer characterization. BCR results showed that Si, Al, Ca, Mg, Fe, and Ba were the predominant inorganic elements in the cake layer, and they occurred mostly as crystal particles. Crystal SiO_2 was the dominant inorganic compound while Ca in the form of $CaSO_4$ (dominant) and $CaCO_3$ were also present, but exerted little effect on the cake layer structure because most of these compounds were deposited as precipitates on the reactor bottom. EDX results indicated that Si and Al accumulated together along the cross-sectional cake layer in the form of Si–Al (SiO_2–Al_2O_3) crystal particles.

1. Introduction

Over the past decade, membrane bioreactors (MBRs) have been used increasingly often in municipal wastewater treatment as a means to increase effluent quality (Meng et al., 2007; Rui et al., 2005; Zhou et al., 2014a). However, membrane fouling can result in decreased membrane performance, reduced productivity and increased costs for cleaning and membrane replacement (Gao et al., 2011; Xia et al., 2008, 2012). Consequently, membrane fouling has become a key challenge for the application of MBRs. In recent studies, the accumulation of a sludge cake layer has been demonstrated to be the major contributor to membrane fouling in MBRs (Guo et al., 2012; Meng et al., 2009; Zhou et al., 2014c).

The deposition of sludge particles, colloids and solutes on the surface of a membrane generates a cake layer, with characteristics

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http://dx.doi.org/10.1016/j.biortech.2015.08.026
0960-8524/

similar to porous media characterized by complex systems of connected inter-particle voids. The cake layer formation is a dynamic process, which can be classified into three general phases: (1) particulate cake initiation due to pore blocking; (2) cake formation due to deposition, accumulation and biological activity; and (3) cake compression (Zhang et al., 2006). Due to the dynamic formation process and the complex conditions within MBR systems, the structure of the cake layer is generally quite complicated. Overall, cake layers can be divided into organic (e.g. polysaccharides, peptidoglycan, proteins, cells and humic aggregates) and inorganic (e.g. colloids and inorganic precipitates such as silicates and calcium sulfate) components. In most studies, the organic components, especially soluble microbial products (SMP) and extracellular polymeric substances (EPS), have been considered the major contributors to cake layer formation and membrane fouling (Guo et al., 2012; Le-Clech et al., 2006; Meng et al., 2011).

The inorganic component of cake layers, which can also play a significant role in fouling, has been described in a variety of studies but has not yet been extensively characterized. For example, Kang et al. (2002) indicated that a thick cake layer composed of biomass and struvite formed on the membrane surface in a membrane-coupled anaerobic bioreactor while Lyko et al. (2007) found that inorganic compounds (mainly metal substances) were more important contributors to membrane fouling than biopolymers. Additionally, recent studies have focused on the fouling variations caused by specific inorganic components (such as Fe^{3+}, Ca^{2+}, Mg^{2+}, Cu^{2+}, CrO^{4-}, etc.), added as amendments in synthetic wastewater containing no insoluble compounds (Arabi and Nakhla, 2009; Katsou et al., 2011; Ognier et al., 2002; Zhang et al., 2008, 2006; Zhou et al., 2014b,c). In addition, Feng et al. (2013) studied fouling variation using municipal wastewater with specific heavy metal addition. Several papers demonstrated that the inorganic components within membrane cake layers are composed of elements such as Mg, Al, Fe, Ca, Si, etc. (Chen et al., 2012; Gao et al., 2011; Meng et al., 2011; Wang et al., 2008; Yang et al., 2011). However, these studies utilized spot-scan energy-dispersive X-ray analyzers (EDX) for cake component analysis, which is only capable of measuring the elemental distribution within a very small spot of the cake layer surface. No suitable method has yet been applied to characterize the detailed inorganic structure and distribution of inorganic components and their chemical structures in cake layers of MBRs treating municipal wastewater.

This study aims to develop an improved fundamental understanding of the structure and distribution of inorganic components in the fouling cake layers of MBR systems used for municipal wastewater treatment. A laboratory-scale submerged anoxic–oxic membrane bioreactor (A/O-MBR) was operated with municipal wastewater influent for over 90 days to characterize the cake layer by various methods. The BCR (European Community Bureau of Reference) sequential extraction, X-ray photoelectron spectroscopy (XPS), and both map scan and line scan of EDX were performed for characterization of the inorganic distribution along a cross-section of the cake layer.

2. Methods

2.1. A/O-MBR setup

For this study, a laboratory-scale A/O-MBR (Fig. 1) with a working volume of 4.5 L (anoxic and oxic zones of 1.5 L and 3.0 L, respectively) was operated for 90 days. A polyvinylidene fluoride hollow fiber membrane module (pore size 0.4 μm) with a total surface area of 260 cm^2 (Litree Company, China) was mounted in the oxic zone and a constant fluid flux was set at 17 L/($m^2 \cdot h$) with an intermittent suction mode (10 min suction and 2 min relaxation for each cycle). Air (0.4 m^3/h) was supplied continuously through a diffuser to provide oxygen for microbial activity and to induce a cross-flow action for effective scouring of the membrane surface. The air flow rate was adjusted with a gas flow-meter and trans-membrane pressure (TMP) was monitored with a pressure gauge.

2.2. Operational condition

The effluent from an aerated grit chamber of the Quyang municipal wastewater treatment plant (WWTP) (Shanghai, China) treating municipal wastewater and storm water from residential areas and construction sites was used as influent to the anoxic MBR zone. Influent characteristics are shown in Table 1 and S1 (Supporting Information). The inoculating biomass was drawn from the return activated sludge stream in the Quyang WWTP. The newly inoculated A/O-MBR was initially operated for 60 days to achieve steady state for the acclimatization of activated sludge. The membrane module was then replaced with a new unit and the A/O-MBR was operated for 90 days for the experiments.

Hydraulic retention time (HRT) and solids retention time (SRT) were maintained at 10.0 h and 30 days, respectively. Mixed liquor suspended solids (MLSS) in the anoxic and oxic zones were approximately 3.8 ± 0.3 and 4.2 ± 0.5 g/L during the experiment, respectively. The flow rate of recycled mixed liquor from the oxic zone to the anoxic zone was controlled at 200% of the influent flow rate.

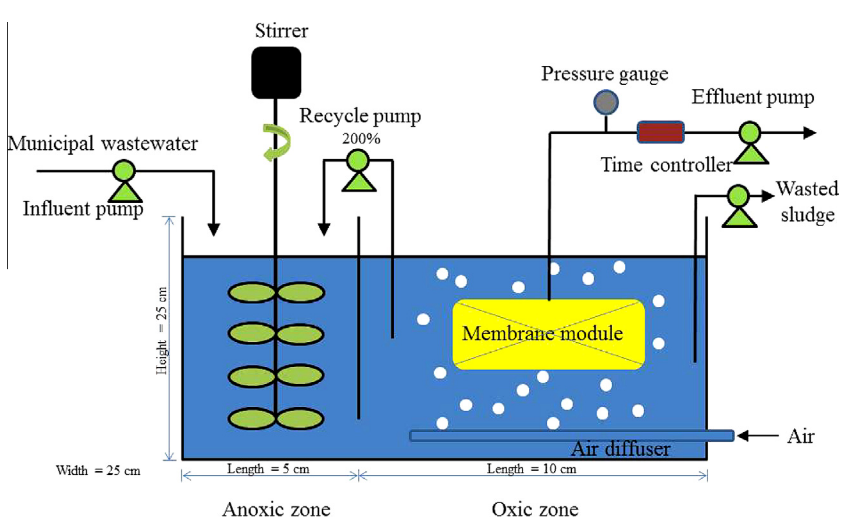

Fig. 1. A schematic of the anoxic/oxic membrane bioreactor used in this study.