Bioresource Technology 171 (2014) 240–246

Contents lists available at ScienceDirect

Bioresource Technology

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

Effect of granular activated carbon addition on the effluent properties and fouling potentials of membrane-coupled expanded granular sludge bed process

An Ding [a], Heng Liang [a,*], Fangshu Qu [a], Langming Bai [a], Guibai Li [a], Huu Hao Ngo [b], Wenshan Guo [b]

[a] State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, 73 Huanghe Road, Nangang District, Harbin 150090, PR China
[b] Faculty of Engineering, University of Technology, P.O. Box 123, Broadway, Sydney, NSW 2007, Australia

HIGHLIGHTS

- Adding GAC alleviated membrane fouling of a membrane-coupled EGSB process.
- It reduced the concentrations of SMP, SMP_{ps} and SMP_{pr} by 26.8%, 27.8% and 24.7%.
- It primarily reduced tryptophan proteins, aromatic proteins and fulvic substances.
- GAC addition mainly decreased the cake layer resistance proportion by 53.5%.

ARTICLE INFO

Article history:
Received 15 July 2014
Received in revised form 14 August 2014
Accepted 17 August 2014
Available online 23 August 2014

Keywords:
Membrane-coupled anaerobic process
Membrane fouling
Granular activated carbon
Membrane resistance
Soluble microbial products

ABSTRACT

To mitigate membrane fouling of membrane-coupled anaerobic process, granular activated carbon (GAC: 50 g/L) was added into an expanded granular sludge bed (EGSB). A short-term ultrafiltration test was investigated for analyzing membrane fouling potential and underlying fouling mechanisms. The results showed that adding GAC into the EGSB not only improved the COD removal efficiency, but also alleviated membrane fouling efficiently because GAC could help to reduce soluble microbial products, polysaccharides and proteins by 26.8%, 27.8% and 24.7%, respectively, compared with the control system. Furthermore, excitation emission matrix (EEM) fluorescence spectroscopy analysis revealed that GAC addition mainly reduced tryptophan protein-like, aromatic protein-like and fulvic-like substances. In addition, the resistance distribution analysis demonstrated that adding GAC primarily decreased the cake layer resistance by 53.5%. The classic filtration mode analysis showed that cake filtration was the major fouling mechanism for membrane-coupled EGSB process regardless of the GAC addition.

1. Introduction

Due to the importance of energy recovery and resources recycling, anaerobic membrane bioreactor (AnMBR) and membrane coupled anaerobic processes have become more and more promising technologies for wastewater treatment in recent years (Stuckey, 2012). Although AnMBR has advantages such as high removal efficiency of organic matters and small footprint, etc., there are still some challenging issues. Particularly, membrane fouling is the key challenge for the widespread applications of AnMBR (Guo et al., 2012; Meng et al., 2009).

It is reported that soluble microbial products (SMP) or loosely bound extracellular polymeric substances (EPS), which are produced from cell metabolism and lysis, play an important role in membrane fouling (Lin et al., 2009; Wang et al., 2014). Barker and Stuckey (1999) have reviewed the advanced treatments such as activated carbon, synthetic resin adsorption, ozonation, oxidation, coagulation and breakpoint chlorination for reducing SMP. Among all the options, granular activated carbon (GAC) is the most effective method for the removal of SMP.

As GAC has high removal efficiency of SMP than others (powdered activated carbon, synthetic resin, etc.) (Barker and Stuckey, 1999), several researchers investigated the GAC addition in the membrane bioreactor (MBR) system to alleviate membrane fouling. Johir et al. have reported that the addition of GAC as a suspended medium in a submerged membrane bioreactor (SMBR) achieved high organic removal (95%) as well as reduced transmembrane pressure (TMP) development by 58%, as GAC addition eliminated the organic molecules in SMP with molecular weight of

* Corresponding author. Tel./fax: +86 451 86283001.
E-mail address: hitliangheng@163.com (H. Liang).

http://dx.doi.org/10.1016/j.biortech.2014.08.080
0960-8524/

1200–150 Dalton (Da) (Johir et al., 2011, 2013). Kim et al. (2010) have also found that GAC addition into an anaerobic fluidized bed membrane bioreactor could reduce membrane fouling rate efficiently.

To date, few studies have explained the mechanisms or given the reasons of GAC alleviating membrane fouling in membrane-coupled anaerobic reactors. Thus, it is necessary to know which kinds of organic matters are easily absorbed and removed by GAC addition and how GAC affect the filtration resistance distribution in membrane-coupled anaerobic reactors system. In this study, the effect of GAC addition on the performance of an expanded granular sludge bed (EGSB) was investigated. The properties of effluents were characterized. The short-term experiments on membrane fouling potentials and mechanisms were carried out in a dead-end ultrafiltration (UF) setup.

2. Methods

2.1. EGSB setup

As shown in Supplementary data, the performance of two lab-scale EGSBs (EGSB1 and EGSB2) fed with synthetic wastewater were examined in parallel during 3 month operation. GAC with the concentration of 50 g/L was added into EGSB1 at the beginning of the operation. The GAC concentration was chosen based on the study of Kim et al. (2010). EGSB2 was a control system without GAC addition. The effective working volume of each EGSB was 3 L (diameter of 50 mm and length of 1800 mm), and the effluent flow rate was set at 0.75 L/h, corresponding to a hydraulic retention time (HRT) of 4 h. Some portion of mixed liquor on the top was recycled back to the reactor with a liquid upflow velocity of 10 m/h. Both of the reactors were inoculated with 10 g/L granular sludge (with the average particle size of 940 μm) originating from a large-scale upflow anaerobic sludge blanket of a soybean wastewater treatment plant in Harbin, China. The granular sludge was washed using deionized (DI) water for 3 times before inoculation.

In order to simulate the domestic sewage, the synthetic wastewater consisted of glucose (200 mg/L), sodium acetate (150 mg/L), NH_4Cl (150 mg/L), KH_2PO_4 (22 mg/L), $NaHCO_3$ (400 mg/L), and a mixture of trace elements. The feed solution contained 310–360 mg/L of chemical oxygen demand (COD), 35–45 mg/L of NH_4^+-N, 4–5 mg/L of TP, and pH value of 7.0–7.5.

2.2. Adsorbents and static adsorption tests

The GAC was made of coconut shell from Tianjin Binhai Kody Company. The particle size of GAC was between 500 and 1700 μm (screen mesh: 10–30 meshes). It had a BET surface area of 867 ± 10 m^2/g.

GAC were washed with DI water for several times until the COD of the supernatant was less than 5 mg/L (limit of detection) before used. GAC static adsorption experiment was carried out in a 1 L baker with magnetic stirrer. GAC with the mass concentration of 50 g/L was added in the baker. The solution stirred at 150 rpm and 25 °C for 24 h. The supernatant samples were taken out for COD analysis at 0 h, 2 h, 4 h, 12 h and 24 h, respectively.

2.3. Short-term UF tests

Bench scale short-term dead-end ultrafiltration (UF) tests were carried out to study the membrane fouling potential of the EGSB effluents during 75–80 days. The effluents samples were collected from the outlets of EGSB1 and EGSB2 (Sample 1 from EGSB1: S1; Sample 2 from EGSB2: S2) for the short-term UF tests. Each UF test was carried out in triplicates.

The filtration system consisted of a nitrogen gas cylinder, an UF cell and an electric balance and a computer (Supplementary data). Flat sheet polyethersulfone (PES) UF membranes (MWCO 100 kDa, OM100076, Pall, USA) with an effective surface area of 43.01 cm^2 were used. The volume of the UF cell (Amicon 8400, Millipore, USA) was 350 mL without stirring. The membrane was placed at the bottom of the cell with glossy side towards the bulk solution. Nitrogen gas was used to drive the feed solution through the membrane at a constant pressure of 30 kPa. The filtrate flowed into a 500 mL beaker on the electronic balance which was connected to a computer. The weighting data were automatically logged every 5 s.

2.4. Membrane resistance model

To evaluate fouling behaviors of the membrane, Darcy's law was applied to estimate the total fouling resistances as shown in Eq. (1):

$$R_t = \frac{\Delta P}{\mu J} \tag{1}$$

where J is the final permeate flux, ΔP is trans-membrane pressure, μ is dynamic viscosity, and R denotes the resistance:

$$R_t = R_m + R_{cp} + R_p + R_c \tag{2}$$

$$R_m = \frac{\Delta P}{\mu J_0} \tag{3}$$

$$R_{cp} = R_t - \frac{\Delta P}{\mu J_1} \tag{4}$$

$$R_c = \frac{\Delta P}{\mu J_1} - \frac{\Delta P}{\mu J_2} \tag{5}$$

As shown in Eq. (2), R_t, R_m, R_{cp}, R_p and R_c are total, membrane, concentration polarization layer, pore blocking and cake layer resistances, respectively (Li and Wang, 2006). R_m is determined by filtering the DI water through the clean membrane; J_0 is the permeate flux of DI water filtered through the clean membrane (from Eq. (3)); R_{cp} is calculated by filtering the DI water through the fouled membrane; and J_1 is the final permeate flux (from Eq. (4)). R_c is determined from the difference in resistance before and after gentle membrane cleaning to remove the cake layer using a sponge, and J_2 is the later flux (from Eq. (5)).

2.5. Modeling for membrane fouling process

The flux decline of UF in the dead-end cell under constant pressure could be described by different blocking mechanisms: complete blocking, standard blocking, intermediate blocking and cake filtration (Shen et al., 2010). The equations for different membrane blocking mechanisms are listed below: (1) Complete blocking: $-J + J_0 = aV$; (2) Standard blocking: $1/t + b = J_0/V$; (3) Intermediate blocking: $-\ln J + \ln J_0 = cV$; (4) Cake filtration: $1/J - 1/J_0 = dV$, where V is the volume of the feed water; a, b, c and d are all constants.

2.6. Analytical methods

COD was measured according to Standard Methods (CEPB, 2002). Turbidity was determined by the Turbidity Meter (HI-98713-02 ISO, HANNA, US). Ultraviolet absorbance at 254 nm (UV_{254}) was determined by a spectrophotometry (SPECORD 50 PLUS, Germany). Particle size distribution was measured using MasterSizer Laser Diffraction Particle Size Analyzer (MasterSizer 2000, Malvern Instruments, England). The SMP was obtained by measuring the dissolved total organic carbon (TOC) in the effluents. The effluent sample was centrifuged at 4000 rpm for 10 min, and then filtered through a 0.45 μm membrane. TOC concentration