Chemical Engineering Journal 279 (2015) 725–734

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Toward energy-neutral wastewater treatment: A membrane combined process of anaerobic digestion and nitritation–anammox for biogas recovery and nitrogen removal

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HIGHLIGHTS

- Combined process was developed by treating concentrated municipal wastewater under ambient temperature.
- The process was suitable for start-up of the anaerobic digestion and nitritation–anammox.
- The process presented a stable removal performance for nitrogen and COD associated with stable methane production.
- The net energy consumption estimated of combined process was 0.09 kWh/m^3.

ARTICLE INFO

Article history:
Received 15 February 2015
Received in revised form 9 May 2015
Accepted 11 May 2015
Available online 19 May 2015

Keywords:
Anaerobic digestion
Mainstream nitritation–anammox
MBR
CANON
Energy-neutral wastewater treatment

ABSTRACT

Energy-neutral wastewater treatment is increasingly emphasized in practice. In this study, a membrane combined process of a submerged anaerobic MBR and CANON MBR was investigated for biogas recovery and nitrogen removal by treating membrane concentrated municipal wastewater under ambient temperatures. Anaerobic digestion and nitritation–anammox were successfully started up in MBRs within a shorter period. During the sequential operation of 81 d, the COD removal efficiency of the whole system was about 96% and most CODs were converted into biogas in the anaerobic MBR with a stable methane yield of 1672 mL/d, corresponding to 223 mL CH_4/g COD. Meanwhile nitrogen removal was obtained in the sequential CANON MBR with a total nitrogen removal efficiency of 81%. Furthermore, the net energy consumption estimated of membrane combined process was 0.09 kWh/m^3. These results indicated that the membrane combined process is a promising treatment process to achieve energy-neutral municipal wastewater treatment.

1. Introduction

Municipal wastewaters are typically treated through conventional activated sludge (CAS) processes which are generally energy intensive and focus on the removal of organics and nutrients. The specific energy consumption of CAS plants without sludge incineration is in the range of 0.3–0.6 kWh/m^3 [1]. About half of the energy is used for aeration, which is necessary for microbial degradation of organic C and nitrogen removal in wastewater. Thus, approximately 45% of the total biodegradable chemical oxygen demand (COD) in wastewater is lost through oxidation to carbon dioxide [2]. Meanwhile, municipal wastewater is considered a valuable resource instead of a waste. This 'used water' is well suited for energy generation, mineral and organic fertilizer production, and clean water production [3–5]. The total energy contents of municipal wastewater embedded in organic C, ammonium-N, and phosphate-P are estimated at approximately 23, 6, and 0.8 w/capita, respectively [3]. Therefore, net energy-neutral or energy-positive municipal wastewater treatment is possible [6].

Several innovative treatment technologies have been developed to achieve energy sustainability. For example, anaerobic digestion (AD) processes enable efficient recovery of energy in the form of methane (CH_4) from wastewater to partially offset the energy consumption for treatment; microbial fuel cell (MFC) enables direct recovery of electric energy and value-added products [7]; coupled anaerobic fermentation and MFC techniques with CAS enables the energy recovery from sewage or sewage sludge [8]. Among of them, the mainstream combination of AD and nitritation–anammox is a potential energy-neutral process. AD is an attractive process for sewage because of its low construction, operation, and maintenance costs; small land requirement; low excess sludge production; and

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http://dx.doi.org/10.1016/j.cej.2015.05.036
1385-8947/

potential biogas production [9]. Similarly, nitritation–anammox allows a significant decrease in the operational costs compared with conventional nitrification/denitrification [4]. However, implementation of both processes remains a challenge. The widespread application of AD for wastewater treatment is limited because of biomass retention, low concentration of wastewater organics, and limitation of operating temperature [10]. For the nitritation–anammox, full-scale implementation at municipal wastewater treatment plants (WWTPs) is constrained to nitrogen removal of sludge digester supernatant in sidestream. The characteristics of digester supernatant facilitate the growth of aerobic ammonium-oxidizing bacteria (AerAOB) and anammox as well as the inhibition of nitrite-oxidizing bacteria (NOB) and heterotrophic denitrifiers [3]. Compared with sidestream anammox, out-selection of NOB and heterotrophic denitrifiers is more challenging in the mainstream, because the ammonium concentration in mainline is not high enough to produce high free ammonium (FA) to suppress NOB growth [11]. Additionally, in the mainstream, high C/N ratios may be favorable to the growth of heterotrophic denitrifiers, whereas low temperature may decrease anammox activity [12]. Consequently, the key to improve the performance of the mainstream combination of AD and nitritation–anammox is to ensure sufficient biomass to offset the slow growth rate of AD and anammox bacteria under ambient temperature. Adopting MBR into the mainline combination of AD and nitritation–anammox can facilitate rapid start up process and high growth rate of slow-growing bacteria with complete biomass retention. As a result, MBR can provide an alternative strategy for low-strength wastewater treatment, with the potential for high biogas recovery and high-quality effluent [13–15]. Meanwhile, raw wastewater should be concentrated to facilitate the application of AD and nitritation–anammox. Direct membrane filtration (DMF) of wastewater has recently gained attention, because it provides the opportunity of concentrating organics and nutrient via membrane concentration; its other advantages include high-quality effluent, simple design, and simple maintenance [16].

In the present study, we explored a membrane combined process of AD and nitritation–anammox, which includes an anaerobic membrane bioreactor (AnMBR) and an MBR applied in completely autotrophic nitrogen removal over nitrite process (CANON). Additionally, a dual membrane device [ultrafiltration (UF)–reverse osmosis (RO)] is used as pretreatment to up-concentrate municipal wastewater for elevating the concentrations of COD and N from municipal wastewater. Thus, in AnMBR, the organic C of concentrate from UF and RO was removed to generate biogas. In CANON MBR, concentrated ammonium was partially oxidized to nitrite by AerAOB, and nitrite and remaining ammonium were converted to nitrogen gas by anammox [17].

This study aimed to investigate the performance of the membrane combined process of AD and nitritation–anammox for membrane concentrated municipal wastewater. The performance of this combined process was evaluated based on (a) biological start-up process on MBR; (b) COD and nitrogen removal efficiency; (c) methane yield; (d) filtration performances of the two MBRs; and (e) energy estimation of the process. With respect to the up-concentration of the dual membrane device, the related research content remains to be published.

2. Materials and methods

2.1. Experimental set-up

The laboratory-scale AnMBR-CANON MBR combined system that was used in this study is shown in Fig. 1. The system consisted of two major containers with an effective volume of 4 L each. One was used as AnMBR, the other was used as CANON MBR. Between the two reactors, a buffer tank was used to collect the AnMBR effluent which was delivered to CANON MBR successively by a peristaltic pump. In both reactors, an identical flat-sheet polyvinylidene fluoride (PVDF) (SINAP, Shanghai, China) membrane module was submerged in the mixed liquor. Nominal pore size of the membranes used was 0.1 μm, and area of the membrane surface immersed was 0.024 m^2. Four peristaltic pumps were individually used to feed influent into the membrane reactors and withdraw permeate from the membrane modules. Membrane fouling was indicated by an increase in the trans-membrane pressure (TMP), which was recorded by a pressure sensor (Endress and Hauser) installed between the membrane module and the permeate pump. Meanwhile, total membrane resistance (R_t) was also calculated to display the membrane fouling. Gas sparging was used to minimize particle deposition on the membrane surface and mix sludge in the reactors. Produced biogas was circulated by a vacuum pump (N86KNE, KNF, Germany) and coarse diffusers were installed at the bottom of the membrane module to generate coarse biogas bubbles in both reactors. During the initial start-up stage of reactors, gas sparging was not used because of less produced biogas. In CANON MBR, three microporous aerators were used to supply air intermittently. Aeration operation was maintained for 2 min on and 3 min off, which was controlled by a programmable logical control (PLC) device. To control the temperatures during the start-up stage, the reactors were jacketed and connected to a water heating system. Water heating was canceled after the start-up stage so that the combined system was operated under ambient temperature conditions. Moreover, a UF-RO dual membrane device (Zhiyuanweiye, China) was used to produce the concentrate. This device consisted of a UF membrane (spiral wound module, PVDF, membrane area of 0.83 m^2) and RO membrane (spiral wound module, aromatic polyamide, membrane area of 1.30 m^2). Both membranes were purchased from Vontron Technology Co. (China).

2.2. Combined system operation

Considering the specific requirements of operation in both reactors, a two-step strategy was developed to rapidly realize continuous operation of the combined system. The first step was the start-up procedure of the two reactors. In CANON MBR, nitrifying sludge of 4.0 g MLSS/L from a municipal WWTP was initially seeded to the reactor for enrichment of AerAOB. Synthetic wastewater, prepared with tap water and containing $(NH_4)_2SO_4$ (N source) and $KHCO_3$ (C source and buffer), was fed to the reactor with pH range of 8.2 ± 0.3. In addition, the synthetic solution contained (per L of tap water) $MgSO_4$, 0.015 g; $CaCl_2$, 0.02 g; K_2HPO_4, 0.028 g and 2 mL of trace elements [18]. During this period (days 1–42), the N loading rate was changed through variations in influent NH_4^+ concentration. Dissolved oxygen (DO) of the reactor was limited to approximately 0.8 mg/L, which was set for ensuring substrate inhibition and DO limitation on NOB [11,19]. The nitrite accumulation ratio (NAR) was calculated using the following equation [20]:

$$\mathrm{NAR} = \frac{[\mathrm{NO_2^-}-\mathrm{N}]_{\mathrm{eff}}}{[\mathrm{NO_2^-}-\mathrm{N}]_{\mathrm{eff}} + [\mathrm{NO_3^-}-\mathrm{N}]_{\mathrm{eff}}} 100\% \qquad (1)$$

Once a stable NAR had been achieved (NAR increased to 75% and more than 50% conversion of NH_4^+), approximately 1.5 L (4.0 g MLSS/L) of anammox biomass from a laboratory-scale up-flow anaerobic sludge blanket reactor treating landfill leachate was added to the reactor (day 36). Subsequently, intermittent aeration was used to realize anoxic condition (DO, 0.3 mg/L), thereby facilitating the coexistence of AerAOB and anammox. From day 36 to day 66, influent NH_4^+ concentration was gradually decreased to