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Feasibility of combined upflow anaerobic sludge blanket-aerobic membrane bioreactor system in treating purified terephthalic acid wastewater and polyimide membrane for biogas purification

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ABSTRACT

Anaerobic processes have become actively practiced for waste and wastewater treatment during the recent years because it can apply for high organic loading whereas the producing biogas can be recovered as a new energy source. This study prompted to look on the feasibility of a combined upflow anaerobic sludge blanket (UASB) and aerated membrane bioreactor (MBR) system for the treatment of purified terephthalic acid (PTA) wastewater and the usage of polyimide membranes for biogas separation. The results revealed that the combined system is robust and flexible in relation to the change in hydraulic retention times (HRTs) and organic loading rates. The overall chemical oxygen demand (COD) removal of 97% with the maximum loading rate of 20 g L^{-1} per day was achieved at 12 h HRT. The biogas from the UASB reactor was mainly methane (>62%). The separation between CO_2 and CH_4 was explored using different concentrations of dense polyimide (PIs) membranes and feed pressure. The results revealed that PIs based membranes possess high potential for CH_4 purification with high selectivity and permeability towards CO_2 than CH_4. The expediency of the proposed treatment system and gas-separating module was seen and thus purified biomethane could be mustered continuously.

1. Introduction

Purified terephthalic acid (PTA) is a crucial material for various petrochemical industries that is consumed in polyester production including polyethylene terephthalate (PET) bottles and textile fibers. Generally, the manufacturing of 1 ton PTA generates 3–10 m^3 wastewater with high chemical oxygen demand (COD) concentration up to 20 kg L^{-1} [1]. Terephthalic acid (TA), *p*-toluic acid (*p*-Tol), *p*-carboxybenzaldehyde (*p*-CBA), phthalic acid (PA) and benzoic acid (BA) are known to be the main pollutants present in this wastewater [1–5]. With increasing PET consumption, the treatment of PTA wastewater and contaminated environments have received broad attention for the preservation of natural ecosystems and protection of the environment. Previous studies indicated that the PTA effluent has potential toxic effects to the living organisms and animals [6,7] and thus it needs to be treated before discharged into the environment.

During the last few decades, various treatment technologies including advanced oxidation process [7–9], anaerobic fermentation [2,3,10–12], aerobic biodegradation [13] and chemical coagulation/flocculation [14] have been investigated for treating organic pollutants present in the petrochemical wastewaters. Among these, anaerobic biodegradation is likely the most cost-effective treatment process with regard to their well known advantages such as energy production via biomethane and lower sludge production and also used for treating high concentration of wastewater. Moreover the energy produced by this method is considered as clean and support to reduce the greenhouse gas emissions. Especially anaerobic treatment by methane fermentation does not require any external addition of oxygen or nitrate as an electron acceptor, the presence of organic matter itself or the carbon dioxide acts an acceptor. Recently, two-stage UASB is considered as the best anaerobic treatment technology for treating PTA wastewater [10,15,16]. Kleerebezem et al. [17] acheived the maximum 25 g L^{-1} d^{-1} volumetric loading rate (VLR) for treating synthetic PTA wastewater with 90% COD removal at 5.6 h HRT whereas long HRT of 96 h was required to reach the same treatment efficiency for real wastewater [2]. According to Kim et al.

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[10] quantity and quality of seeded biomass also plays an important role for acheiving high treatment efficiency. Long sludge acclimatization time is normally required in order to enrich TA-degrading biomass [10,12]. Effect of other operating parameters such as HRT, VLR, pH and temperature are also intensively described in the literatures [2,3,10–12].

Although the anaerobic process is reliable in removing organic pollutants from PTA wastewater, however the disadvantages like high COD and pathogens concentration were observed in the effluent which decreases the process efficiency. In order to comply with new discharge standards and to have better quality of treated wastewater, the combined anaerobic-aerobic treatment processes are commonly preferred [11,18]. In aerobic process, membrane bioreactors (MBRs) show a better process performance as compared to the conventional activated sludge. The reactor is developed by combining aerobic biodegradation and membrane filtration in a single reactor, allowing more flexibility in the application of different modification. Moreover, long sludge retention time (SRT) has made this technology more attractive for treatment of recalcitrant wastewater [18,19]. Thus, implementation of MBR as a post treatment of anaerobic process could achieve high quality of treated effluent, which might give possibilities for wastewater reuse.

In addition to the wastewater treatment, biogas production and its separation are considered to be important because the produced purified methane can be used for onsite heating or electricity generation. Removal of gases such as CO_2, H_2S, H_2 and water from anaerobic digestion can be performed through adsorption, chemical and membrane separation which can enrich the production of methane [20]. Due to the development of new polymer materials with higher performance in term of permeability and selectivity, application of membranes for gas separation has become more feasible from economic and technological point of view [21,22].

To our knowledge, there is no research exists that focuses on treatment of PTA wastewater using a combined process of UASB-MBR system and biogas separation using membrane. In this work, we first investigated the performance of UASB and MBR for COD removal at different HRTs and loading rate followed by the fabrication of polyimide membrane for separation of methane from the mixed biogas. Effect of polyimide concentration and feed pressure on permeability and selectivity were also observed.

2. Materials and methods

2.1. Experimental setup of the combined UASB-MBR system

Fig. 1 shows a flow diagram of lab-scale combined UASB-MBR system. The UASB was made of transparent acrylic cylinder, with 150 mm internal diameter, 1500 mm height and a working volume of 25 L. The influent and effluent flow rates were controlled by variable-speed peristaltic pump. The biogas produced from the reactor was separated from anaerobic biomass and the effluent by a gas-liquid-solid separator equipped at 50 mm below the effluent port. A temperature of 35 °C was controlled throughout the experiment by recirculating hot water outside the reactor wall. The reactor pH was manually maintained in a range of 6.5–8.5 using 0.1 N $NaHCO_3$ and 0.4 N HCl solutions when necessary.

The aerobic MBR was operated using the anaerobic effluent. The reactor was rectangular tank of 300 mm length, 185 mm width and 450 mm height, having the effective volume of 25 L. A submerged cross-flow polyvinylidene difluoride (PVDF) flat sheet membrane module with pore size of 0.45 μm were placed inside the reactor and air diffuser with the rate of aeration is 4 L L^{-1} min was equipped under the membrane module to reduce the membrane fouling and also to maintain an aerobic environment in the reactor. During the entire experimental period, the reactor was operated with the membrane flux ranging between 2.5–3.2 L M^{-2} h^{-1}. No excess sludge was drained in both the UASB and the MBR reactors throughout the experiment.

2.1.1. Reactor operation

Inoculum sludge for the UASB and MBR was obtained from anaerobic and activated sludge treatment units of a petrochemical company and domestic wastewater treatment plant located in Taoyuan County, Taiwan. Anaerobic biomass was acclimatized with PTA effluent obtained from a local petrochemical industry for 100 days. During the experiment, $NaHCO_3$ was provided to keep

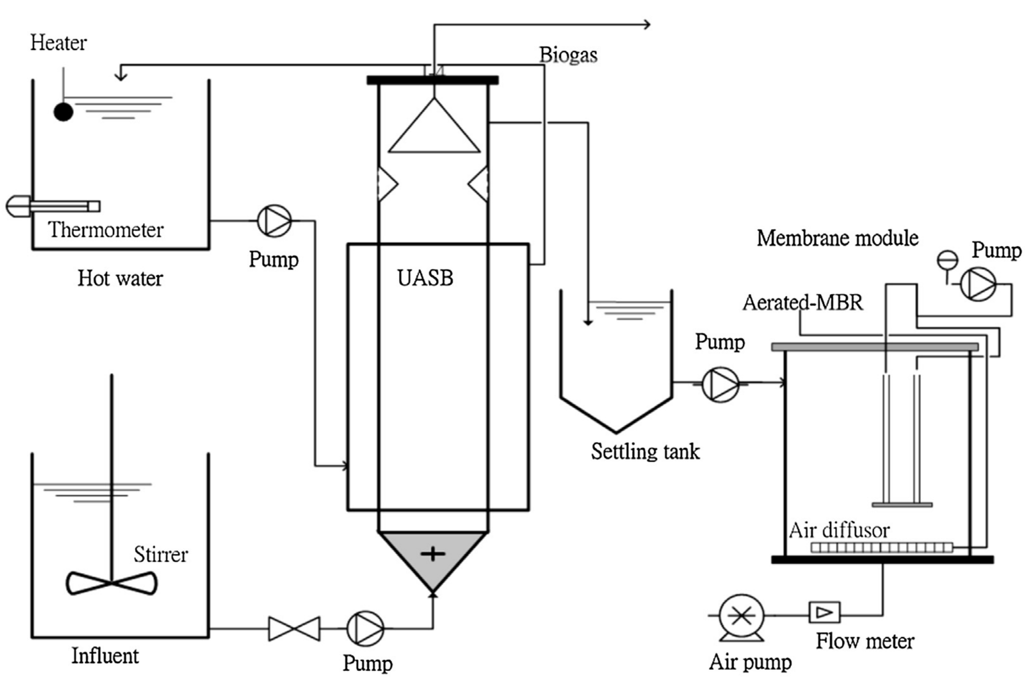

Fig. 1. Schematic diagram of lab-scale combined UASB –MBR system.