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Comparative study of degassing membrane modules for the removal of methane from Expanded Granular Sludge Bed anaerobic reactor effluent

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ABSTRACT

The feasibility of an emergent technology for in situ removal/recovery of methane from the effluent of an Expanded Granular Sludge Bed (EGSB) anaerobic reactor has been studied. For this purpose, the performances of two commercial hollow fibre degassing contactors with different membrane materials – microporous (polypropylene, PP) and non-porous (polydimethylsiloxane, PDMS) – were compared. The influence of water fluxes ($Q_L/A_{membrane}$ ranging from 22.6 to 377.4 L h^{-1} m^{-2}), vacuum pressure (140–800 mbar), sweep gas fluxes ($Q_{N2}/A_{membrane}$ ranging from 0.14×10^3 to 4.44×10^3 L h^{-1} m^{-2}), and mode of operation (liquid flowing in the lumen side or the shell side) was studied. Both materials showed different behaviours with the variations in operational conditions. In liquid flowing in the lumen mode operation, PP microporous membrane was slightly more efficient under soft or mild operational conditions (low liquid flow and/or vacuum pressure) but showed a wetting phenomenon when operational conditions were harder. In shell side mode, PDMS was more efficient and no wetting phenomenon was observed with this contactor. The differences have been explained, taking into account the material properties (porosity, material resistance ...) of the membrane and structure (packing density, fibre diameter ...) of the modules. Methane removal efficiencies of up to 98% could be achieved, showing the viability of methane removal/recovery using this technology. Simultaneous degassing of CO_2 was also monitored in both modules, showing that the removal efficiency of this gas was considerably lower than for methane. In general terms, the removal of dissolved CO_2 followed a quite similar behaviour from that described for methane. Experimental overall mass transfer coefficients were also obtained.

1. Introduction

Anaerobic wastewater treatment is a widely used technology for industrial wastewaters. One of the advantages of the anaerobic treatment, compared to aerobic systems, is that the process produces biogas, which can be used as fuel for the generation of electricity or heat for domestic and industrial use. Raw biogas is primarily methane and carbon dioxide, and may have small amounts of hydrogen sulphide, moisture, and siloxanes. Depending on organic compounds in treated water, biogas can have different composition. The percentage of methane in biogas typically varies between 50% and 75%. This value can be even higher depending on interaction with aqueous phase of the carbon dioxide. In addition, anaerobic treatment presents important benefits such as lower production of solids, lower requirements for nutrients, lower energy requirements, and a smaller required volume (higher organic loads) than most conventional biological treatments [1].

Domestic and various industrial wastewaters, such as those from the malting industry, bottling processes, drink manufacturing plants, and breweries, are conventionally discharged at moderate to low temperature. The conventional mesophilic anaerobic treatment of such wastewaters (35–40 °C) implies the heating of the reactor content in a more complex system with extra energy consumption from the biogas produced. In some industrial applications, these drawbacks limited the application of this treatment. In this context, some investigations have focused on the study of anaerobic treatment at low temperatures (psychrophilic conditions) for different industrial wastewaters [2–6]. The feasibility of this technology is nowadays proven, and in some cases it can be considered as a convenient option. Nevertheless, low wastewater temperature processes involve a significant quantity of residual methane present in the water effluent of the reactor, as methane solubility rises with the decrease of temperature. The recovery of the residual methane in the liquid anaerobic effluent (R-CH_4) is important for several reasons. Firstly, R-CH_4 discharge of these kinds of effluents represents a loss of a potential energy source. In addition, emissions with R-CH_4 can also generate explosive

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atmospheres when the effluent is discharged to drain or into other closed containers, so it is important to adhere to the safety requirement of keeping the methane concentration in air below the Lower Explosive Limit (5% v/v). Finally, R-CH_4 discharge causes an important carbon footprint to the environment due to fugitive emissions of CH_4 [7,8], as the global warming potential of methane is 28 times higher than that of carbon dioxide [9].

The removal of dissolved gases from liquids is conventionally achieved with vacuum packed towers. These columns are filled with packing that creates a large surface area for the contact of liquid and gas phases. Nevertheless, in this system, the direct contact of liquid and gas phases can frequently lead to problems such as foaming, flooding, and emulsions. Among the alternative technologies under investigation for the removal of dissolved gas from anaerobic effluents, one can find methods such as micro-aeration using biogas containing air or biological oxidation. Nevertheless, these methods also present drawbacks such as low concentration in the removed gas and/or low recovery efficiency [10–12]. It seems that improvement of the existing process, development of new processes, or both are needed in order to minimize the discharge of R-CH_4.

In this context, degassing membrane (DM) contactors have appeared as an emerging technology that is being used to remove dissolved gases in several processes. The main advantage of this technology is related to the fact that a gas and a liquid phase come into contact in the pore of the membrane, without the need for dispersion of one phase into another, allowing previously mentioned problems to be avoided [13]. DM contactors present other advantages over conventional dispersed phase contactors, such as availability at high and low flow rates as they are modular, ease of scaling up, a wide range of capacities by adding or removing membrane modules, a high interfacial area per volume unit, and high efficiency. Nevertheless, DM contactors can also have some disadvantages such as membrane resistance to mass transfer, bypassing in the shell side, fouling problems, and limitations with regard to pressure drop [14], so investigations like the one presented in this work are still necessary.

A hollow fibre module is the most common configuration used in DM contactors in order to remove gases such as CO_2 and O_2 from a liquid phase, so a considerable number of studies related to the removal or recovery of these gases can be found [15–21]. This technology seems especially interesting in some industrial processes where the removal of dissolved gases is crucial, such as in the process of production of ultrapure water, in which one of the major contaminants is the dissolved oxygen, whose removal is essential [15,19]. Unfortunately, studies on the removal of R-CH_4 from anaerobic effluents are still very scarce. Bandara et al. studied R-CH_4 recovery by degasification from the effluent of a bench-scale upflow anaerobic sludge blanket (UASB) reactor treating synthetic wastewater [22]. They used a multi-layered composite hollow fibre degassing module made of polyethylene and polyurethane under vacuum pressure. Cookney et al. studied the recovery of R-CH_4 from a low-temperature (16 °C) anaerobic process treating domestic wastewater with a polydimethylsiloxane membrane contactor using nitrogen as sweep gas [23]. Recently, Cookney et al. studied the desorption of R-CH_4 from both synthetic and real anaerobic effluents using different membrane hollow fibres [24]. Further research in this field is needed to improve and deepen the knowledge and performance of this technology.

The selection of a suitable membrane is a crucial factor for optimal contactor performance, since membrane material properties can significantly affect the overall mass transfer. In this sense, the increase in porosity of the polymeric material for contactor devices applied to water treatment can have a positive influence on the permeability of dissolved gas but a negative effect on the flooding prevention of the membrane material [25].

The main objective of this work was to investigate the performance of two DM contactors in the recovery of R-CH_4 from a recirculating stream of a lab-scale Expanded Granular Sludge Bed (EGSB) anaerobic reactor. Two different materials for the membrane of the hollow fibre membrane modules are compared: polypropylene (PP, microporous) and polydimethylsiloxane (PDMS, non-porous). The effects of liquid flux, vacuum pressure, sweep gas flow rate, and operating mode (lumen and shell sides) were investigated. The removal of dissolved carbon dioxide (D-CO_2) was also monitored to study the simultaneous removal of both gases.

2. Experimental

2.1. Degassing membrane modules

This study employed two types of hollow fibre membrane contactors, which were selected as representative of two different and efficient types of commercial modules for industrial applications with different porosity properties. The first module was PDMSXA-250, a membrane contactor with fibres of polydimethylsiloxane (PDMS, non-porous) with an internal surface area (A_i) of 0.0159 m^2, supplied by PermSelect, MedArray Inc. (USA). The second module was the 1 × 5.5 MiniModule supplied by Liqui-Cel, Membrana GmbH (Germany). This contactor was made of polypropylene (PP, microporous) fibres with an A_i of 0.18 m^2. The main characteristics of both modules are summarized in Table 1.

Since both modules have different physical properties, including different sizes, in order to compare behaviours and performances between the two contactors, the characteristic parameter defined as the water flux rate (Q_L/A_i, liquid flow rate per the membrane surface, $L\,h^{-1}\,m^{-2}$) has been widely used in this study. This parameter has been selected as the most representative of this type of process especially considering the potential scale-up applications, since the flow rate is the only operational variable that can be modified in a real process and the membrane surface area is commonly used as the representative variable in capital cost evaluation of membrane equipment [26,27].

2.2. Experimental setup and procedure

A laboratory-scale EGSB anaerobic reactor was operated at 25 °C for more than 24 months. The EGSB reactor was initially inoculated with 4 L of granular anaerobic sludge from the wastewater treatment plant of a local brewery. The reactor treated 8 $L\,d^{-1}$ of synthetic wastewater polluted with ethanol with an organic load rate of 32 kg chemical oxygen demand (COD) $m^{-3}\,d^{-1}$. A high recirculation flow was kept to expand the sludge bed with an upflow velocity of 10.7 $m\,h^{-1}$. Similar work conditions were used in the study of Lafita [28]. A liquid–gas separator device was placed at the top of the reactor. The gas outlet was connected to a

Table 1
Characteristics of PDMS and PP modules.

	PDMS	PP
Number of fibres	320	2300
Effective length, m	0.083	0.1397
Inner diameter, μm	190	220
Outer diameter, μm	300	300
Pore diameter, μm	Non-porous	0.04
Internal area (A_i), m^2	0.0159	0.180
External area (A_e), m^2	0.0250	0.303
Shell tube inner diameter, m	0.016	0.025
Packing fraction	0.113	0.33
Maximum flow rate (Q_L), $L\,h^{-1}$	12	30
N_2 flow rate, $L\,h^{-1}$	2.7–27.0	26.0–800