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## Carbon dioxide rich microbubble acceleration of biogas production in anaerobic digestion



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#### HIGHLIGHTS

#### G R A P H I C A L A B S T R A C T

- Sparging pure nitrogen has negative effect on the production of methane in AD.
- Recirculation of biogas in anaerobic digestion can enhance production of CH<sub>4</sub>.
- The methane production rate increases with pure CO<sub>2</sub> in microbubble sparging.

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#### 1. Introduction

Renewable fuels have become the main focus for many researchers interested in the production of sustainable energy. Alternative clean sources of energy are available, for instance, solar, hydroelectric, wind and bio-fuels such as bio-diesel and bio-

Cumulative methane production from the gaslift digester and conventional digester when the pure carbon dioxide is sparged

#### ABSTRACT

This paper addresses the use of anaerobic bacteria to convert carbon dioxide to biomethane as part of the biodegradation process of organic waste. The current study utilises gaslift bioreactors with microbubbles generated by fluidic oscillation to strip the methane produced in the gaslift bioreactor. Removal of methane makes its formation thermodynamically more favourable. In addition, intermittent sparging of microbubbles can prevent thermal stratification, maintain uniformity of the pH and increase the intimate contact between the feed and microbial culture with lower energy requirements than traditional mixing. A gaslift bioreactor with microbubble sparging has been implemented experimentally, using a range of carrier gas, culminating in pure carbon dioxide, in the anaerobic digestion process. The results obtained from the experiments show that the methane production rate is approximately doubled with pure carbon dioxide as the carrier gas for intermittent microbubble sparging.

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ethanol from agricultural crops, waste or microalgae. None of these sources, however, have so far been able to produce sufficient energy to provide a substitute for fossil fuels (Schenk et al., 2008; Singh, 2012; Chisti, 2007; Eriksen, 2008; Kadam, 1997).

Anaerobic digestion represents a renewable energy source (Budzianowski, 2012; Wang et al., 1999). It is commonly used for nutrient and energy recovery from biomass and also to stabilise the sludge produced in wastewater treatment (Tiehm et al., 2001). Organic matter is broken down through four biodegradation stages into methane ( $CH_4$ ), carbon dioxide ( $CO_2$ ), varying amounts

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of hydrogen sulphide (H<sub>2</sub>S), and the digested sludge, which can be used as a soil fertiliser (Poeschl et al., 2010; Budzianowski, 2012). Bio-methane can be used for the generation of electricity or used as a biofuel for vehicles after upgrading processes. The production and upgrading costs of biogas are lower than the costs of production and upgrading of bio-fuel produced from agriculture crops or from microalgae (Appels et al. 2008; Sahlström, 2003; Ahring, 2003; Metcalf and Eddy, 2003). However the challenges facing anaerobic digestion implementation have become a major obstacle to this source becoming a leading renewable energy source. Among these challenges are the low volumetric yields of biogas and difficulties relating to the stability of large-scale continuous operation (Salomoni and Petazzoni, 2006; Metcalf and Eddy, 2003).

This paper introduces the premise of using a microbubble sparging system in anaerobic digestion (AD) primarily to extract methane from the bioreactor. Methane has a low solubility in water and therefore is likely to adhere to the organic phase biomass and microbial membranes. The typical exit route for methane from an AD reactor is to build up a gas layer on the organic phase until sufficient volume is created that buoyant forces detach a large bubble, which is in equilibrium with the aqueous phase due to the long contact time. In this paper, we report on experiments that periodically sparge with a bubble size distribution that includes sub  $100\,\mu m$  size microbubbles. Such microbubbles have a terminal rise velocity  $10^{-3}$  m/s or less, and as shown in Al-Mashhadani et al. (2015a), are readily entrained and therefore have a long residence time - minutes rather than seconds. These circulating microbubbles provide local gas-liquid interfaces which can interact with the methane-rich boundary layers of the organic phase to provide an exit route from the system. Hypothetically, the build-up of methane rich boundary layers surrounding microorganisms could serve as an inhibitor to their metabolism in accordance with Le Chatelier's principle. Such thermodynamic principles are important in anaerobic processes such as those considered in this work which operate close to chemical equilibrium (Hoh and Cord-Ruwisch, 1996). Reducing the chemical activity of the product gases in solution (or the fugacity in the gaseous phase) leads to a negative change in Gibbs free energy. Hence the reaction becomes thermodynamically favourable and provides impetus for the formation of more products. We will describe the chemical potential non-equilibrium thermodynamic drivers underpinning the hypothesis in Section 2; methods and materials in Section 3; and the results in Section 4. Our conclusions will be presented in Section 5.

#### 2. Hypothesis of present study

Sparging of anaerobic digestors will affect the dissolved concentrations of gaseous species such as  $CH_4$ ,  $CO_2$ ,  $H_2$ ,  $NH_3$ ,  $H_2S$ . Since all of these gases are produced by anaerobic digestion of biomass, the most common effect will be for sparging to reduce levels of these species by a stripping effect. This would certainly be the case for sparging with an inert carrier gas such as  $N_2$ . On the other hand, if sparging is carried out with sufficiently high partial pressures of a gas that is produced during anaerobic digestion, there may be a driving force for this species to enter solution thereby increasing its dissolved concentration. If we restrict ourselves to the key species involved in anaerobic carbon catabolism, Note that we are neglecting any other gaseous products or intermediates, most notably  $NH_3$  and  $H_2S$ .

The mathematical relationship between Gibbs free energy and species partial pressure is as follows:

$$CH_3CH_2COOH + 2H_2O \rightarrow CH_3COOH + CO_2 + 3H_2$$
(1)

$$\Delta G = \Delta G^{\circ} + RT \ln \frac{[CH_3CH_2CH_2COOH][CO_2][H_2O]^3}{[CH_3COOH][H_2O]^2}$$
(2)

Where  $\Delta G$  is the Gibbs free energy change,  $\Delta G^{\circ}$  is the standard Gibbs free energy, *R* is universal gas constant, *T* is temperature of reaction.

From the above equation, it is possible to note that decreasing the partial pressure of the products contributes negatively to the Gibbs free energy, hence the reaction becomes thermodynamically favourable towards the formation of more products, and vice versa (Gary, 2004). Biogases produced by AD can be either present in a gaseous film or dissolved in the bulk liquid as Eqs. (1) and (2) are completely general. For ideal gases, the partial pressure is equal to the fugacity from which the chemical potential and species activity can readily be computed.

In biological processes, some required reactions are not spontaneous – i.e. they are thermodynamically unfavourable  $(+\Delta G)$ . Typically, these reactions are driven forward by one of two mechanisms as described below.

The first mechanism employed in metabolic networks is to provide enough energy to endergonic reactions to convert them to spontaneous reactions. Reducing the partial pressure (chemical potential) of products by their removal is another method that can be used to make reactions spontaneous in bioprocesses sharing intermediates. This principle underlies reactive separation that is a staple chemical engineering approach to intensify reactions. For example, fermentation of acetate in anaerobic digestion has a positive standard Gibbs free energy and this reaction shown in Eq. (3) is, therefore, thermodynamically not favoured unless the partial pressure of hydrogen can be reduced by methanogenic bacteria to sufficiently low levels such as  $10^{-4}$  atm.

$$CH_{3}CH_{2}CH_{2}COOH + 2H_{2}O \rightarrow 2CH_{3}COOH + 2H_{2} \Delta G^{0}$$
$$= + 48.1 \text{ kJ/mole}$$
(3)

There has been much investigation of the mathematical relationship between partial pressure and Gibbs free energy with widespread applications. But the major results have emerged from biological processes, particularly for bio-hydrogen production. This process has caused debate among researchers about how to control the partial pressure of hydrogen or carbon dioxide and its effects on the production of hydrogen. Many researchers have noted that an increase in hydrogen production could be achieved by reducing the partial pressure of hydrogen or carbon dioxide or both depending on the following equation:

$$C_6H_{12}O_6 + 2H_2OBacteria2CH_3COOH + 2CO_2 + 4H_2$$
 (4)

Tanisho et al. (1998), Park et al., (2005) and Alshiyab et al., (2008) studied the effects of the reduction of the partial pressure of carbon dioxide on hydrogen production. Tanisho et al. (1998) found that hydrogen production increased when the partial pressure of carbon dioxide decreased. Park et al. (2005) demonstrated that reducing the concentration of carbon dioxide from 24.5% to 5.3% in the headspace caused an increase in the hydrogen yield of 43%. Alshiyab et al. (2008) indicated that there was an increase in the hydrogen yield when partial pressure of carbon dioxide was decreased. Moreover, Liang et al. (2002), Mizuno et al., (2000), Kim et al. (2006) and Kraemer and Bagley (2008) all reported that reducing the partial pressure of hydrogen caused an increase in hydrogen production rate. These investigations have shown the importance of the removal of gases from biological processes and the effect this has on increasing production of hydrogen.

Similarly, for anaerobic digestion, the removal of some gases

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