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Microaerobic control of biogas sulphide content during sewage sludge digestion by using biogas production and hydrogen sulphide concentration

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HIGHLIGHTS

- Microaerobic conditions are applied to controlling the biogas sulphide content.
- Biogas production is used to regulate the oxygen supply.
- Biogas sulphide content is used to optimise the oxygen supply by a PID controller.
- Biogas production can be an efficient regulating parameter at steady sulphur load.
- Biogas sulphide content can be a precise regulating parameter in all conditions.

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ABSTRACT

This paper presents the potentials of using biogas production and hydrogen sulphide concentration as the parameters to regulate the oxygen supply to microaerobic reactors in order to control the biogas sulphide content. Research was carried out in two identical bioreactors of 200 L at 35 °C and 19 d of hydraulic retention time. The feed consisted of mixed sludge from a municipal wastewater treatment plant with variable organic and sulphur load. The oxygen flow rate was automatically adjusted according to the biogas sulphide content (which ranged from 0.62 to 0.24%v/v) by a feedback Proportional-Integral-Derivative controller. The target hydrogen sulphide concentration (0.01%v/v) was achieved in 4.0–5.5 h. The micro-oxygenation level reached was considered to be the optimum in the short-medium term, since it kept the removal efficiency above 99% and minimised the oxygen concentration in the biogas during the days following the controller application. Specifically, the average biogas oxygen content was 0.09%v/v. Subsequently, biogas production was used as the parameter to regulate the oxygen supply. When the biogas sulphide content was around 0.33 and 0.50%v/v, approximately 3.5 and 5.0 NL of oxygen were supplied per N m^3 of biogas (respectively). An average sulphide removal efficiency of 99%, and oxygen concentrations in the biogas of less than 0.08%v/v were achieved. Biogas production could be employed to develop precise control strategies during microaerobic digestion under variable organic load and steady sulphur load. Under unstable sulphur load, biogas sulphide content should be used instead.

1. Introduction

Biogas is a versatile and renewable energy source produced mainly by the anaerobic digestion of sewage sludge [1]. Methane and carbon dioxide are the main constituents, but it also contains significant quantities of undesirable compounds such as hydrogen sulphide, whose concentration can reach 1.0%v/v [2]. It is produced by sulphate-reducing and acidogenic bacteria mainly from sulphate and proteins, respectively [3]. Hydrogen sulphide escapes with the biogas, and has detrimental impacts on society and health, environment, and installations for biogas utilisation. Namely, it causes bad odours and eye damage when values are below 0.01%v/v, and even death when above 0.03%v/v [4]. Manufacturers of combined heat and power (CHP) production units recommend limiting values to between 0.01 and 0.03%v/v in order to prevent corrosion in piping systems and equipment. However, short peaks can occasionally be accepted [5]. Therefore, biogas

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sulphide content has to be controlled in order to prevent damage and fulfil the quality standards required in consistence with the chosen application of the biogas.

Recently, there has been wide interest in desulphurisation biotechnologies, since they are an effective and environmentally friendly way of solving the problems of large investments and operational costs incurred by physicochemical processes [6]. Inside them, hydrogen sulphide is removed by sulphide-oxidising bacteria (SOB), which obtain energy by employing sulphide as the electron donor and oxygen as the electron acceptor [7]. The pathway of sulphide biological oxidation inside bioreactors has been suggested as: $H_2S \rightarrow S^0 \rightarrow S_2O_3^{2-} \rightarrow S_4O_6^{2-} \rightarrow S_3O_6^{2-} \rightarrow SO_3^{2-} \rightarrow SO_4^{2-}$ [8]. Sulphide can be also chemically oxidised [9]. Among the bioprocesses, many investigators have turned to microaerobic removal, which consists of supplying limited amounts of oxygen (or air) directly into the anaerobic bioreactor. This is possible because SOB are present in numerous substrates treated by anaerobic digestion [2]. Thus, no additional unit (such as a bioscrubber, biofilter or biotrickling filter) is needed. It is important to note that these microaerobic reactors yield similar to the anaerobic ones [10], or even better [11].

During digestion under microaerobic conditions, SOB colonise the headspace of the reactor and oxidise hydrogen sulphide by using the oxygen that reaches this area, independently of both the oxygen dosing point and mixing method [12]. Díaz et al. [12] demonstrated that the most efficient reactor configuration in order to microaerobically desulphurise biogas involves injecting the oxidant agent into the headspace and implementing liquid recirculation as the mixing method. Thus, the oxygen consumption in undesired processes was minimised. As a result, elemental sulphur accumulates all over the gas space [13]; it must be taken into account that both reactants (air or oxygen) are supplied in limited amounts in order to minimise both their concentration in the biogas, and the operating costs. With regard to this, it is worth noting that the mixtures of methane–oxygen formed inside microaerobic reactors are far from being explosive, since the limits of methane flammability in air are 5.0–15.0%v/v. Additionally, it should be considered that gases such as carbon dioxide reduce this concentration range [4].

The digester response to the presence of limited amounts of oxygen in terms of biogas composition and, more specifically, in terms of hydrogen sulphide and oxygen concentration, has been proved to be really rapid [14]. Moreover, Jenicek et al. [15] and Díaz et al. [16] highlighted the steady dynamic behaviour of microaerobic reactors treating solid wastes and sewage sludge, respectively, within a wide range of hydrogen sulphide concentrations. They also pointed out that microaeration decreases the heating value of the biogas (that is, methane concentration) due to the presence of nitrogen. Accordingly, though it is expensive, the most profitable oxidant agent is pure oxygen. Nonetheless, it must be taken into account that not all the oxygen supplied to a digester is often used therein [16,17]. With regard to this, it is worth noting that unless the biogas is used for CHP or boilers, the presence of oxygen should be avoided, since it is expensive to remove [18]. In Europe, if biogas is to be used as a vehicle fuel or injected into fuel cells or natural gas networks, its oxygen content must not exceed concentrations of 1.0 and 3.0%v/v, respectively [5].

On a full-scale, since hydrogen sulphide production can vary according to the feeding composition, the micro-oxygenation rate must be periodically regulated in order to avoid a lack or surplus of oxygen in the biogas, while maintaining the quality standards required. For this purpose, the key issue is to find a variable capable of providing a precise control of the oxygen supply; little research efforts have been made within this context. Oxidation–reduction potential (ORP) has been reported as an accurate regulation parameter of oxygen dosing in order to eliminate sulphide toxicity [19], maximise sulphur recovery [20], and even desulphurise biogas [21]. However, its response to micro-oxygenation can be insufficient to develop a reliable control of the hydrogen sulphide concentration in biogas during digestion [16]. On the basis that the gaseous sulphide and biogas production both increase and decrease concurrently with the organic loading rate (OLR) due to rises and decreases in fermentative activity, Díaz et al. [22] proposed to regulate the oxygen flow rate according to the biogas production. They found a linear correlation between the ratio of the oxygen supply to the biogas flow rate, and the biogas sulphide content. Under steady hydrogen sulphide concentration, biogas production would be used to develop a reliable and consistent control strategy. Otherwise, the biogas sulphide content could be an efficient regulating parameter of the micro-oxygenation.

The performance of oxygen utilisation inside a bioreactor is expected to vary with time due to increasing elemental sulphur accumulation in the headspace. This could alter the oxygen transfer conditions and affect biological oxidation rates as a result of the change in the growing conditions. Therefore, a control approach using hydrogen sulphide concentration in biogas as the regulating parameter of the oxygen supply would automatically absorb changes in both performance of oxygen utilisation in the digester and biogas production. Proportional-Integral-Derivative (PID) control is the standard automatic controller in industrial settings. Among this type of control systems, the feedback PID controller determines an input variable to the control process by using the measurement of an output variable [23]. Besides being applicable to many real-world control problems, the PID controller is simple, intuitive, efficient, and reliable for processes with steady dynamic behaviour [24]. Therefore, it could be successfully applied to control the biogas sulphide content in microaerobic digesters by using biogas sulphide content as the regulating parameter of the oxygen flow rate.

The aim of this study is to investigate the feasibility of using biogas production and hydrogen sulphide concentration in biogas to regulate the oxygen flow rate, thereby achieving a consistent and efficient control of the hydrogen sulphide concentration during microaerobic digestion.

2. Materials and methods

2.1. Pilot-scale digesters

Two identical continuous stirred tank reactors called R1 and R2 with a working volume of 200 L and a headspace of 50 L were operated under mesophilic conditions and 19 ± 1 d of hydraulic retention time (Fig. 1). Temperature was maintained by an electric resistor. The pressure control was performed hydraulically; an electro-valve regulated the biogas outflow.

Mixed sludge with variable composition was transported weekly to the pilot plant from a municipal wastewater treatment plant. It was stored at 4 °C, and was fed continuously from two stirred tanks at room temperature into both digesters by peristaltic pumps. Sludge recirculation at a rate of 50 L/h ensured mixing. Pure oxygen from a cylinder was injected by means of two mass flow controllers (Bronkhorst EL-FLOW Select) into the headspace or the recirculation stream, depending on the operational stage (Fig. 1).

2.2. Monitoring and experimental analysis

Digestion pressure and temperature were monitored. Biogas was quantified by the displacement of a fixed liquid volume, and its composition was determined by a CP-4900 Micro-GC [12]. The detection limit (DL) for hydrogen sulphide was 0.001%v/v.

The feed and the digested sludge were sampled weekly for total and soluble chemical oxygen demand, total solids, volatile solids