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Two-stage anaerobic fermentation process for bio-hydrogen and bio-methane production from pre-treated organic wastes

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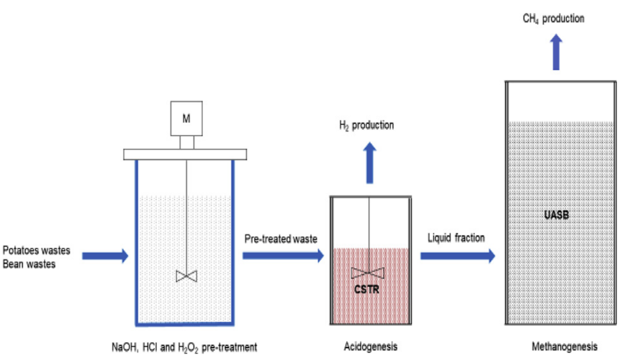
GRAPHICAL ABSTRACT



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ABSTRACT

In this study, the effect of pre-treatments including alkaline, acid and hydrogen peroxide on continuous hydrogen and methane production was investigated. Two different substrates as potatoes and bean wastes were used. Pre-treatment showed positive effect on bio-hydrogen and bio-methane production; higher bio-hydrogen and bio-methane production results using pre-treated samples than the control bioreactors (without pre-treatment), were recorded. In case of potatoes wastes, the hydrogen yield ranged between 126.4 and 252.7 mL-H_2/g-TVS using pre-treated samples compared to 58.7 mL-H_2/g-TVS observed in the reference test. Pre-treated bean wastes showed hydrogen yield of 93.0–152.1 mL-H_2/g-TVS higher than 53.3 mL-H_2/g-TVS measured in the control test. In the second stage, average methane yield results of 322.9–507.1 and 284.3–462.6 mL-CH_4/g-TVS higher than 198.6 and 124.3 mL-CH_4/g-TVS measured for potatoes and bean wastes control bioreactors, respectively. The best results were observed using H_2O_2 pre-treatment. The energy production efficiency was improved by combining H_2 and CH_4 bioreactors.

1. Introduction

Depletion of the energy sources and increasing the environmental pollution due to the excessive use of fossil fuels can be considered as one of the biggest challenges in the future (Akinbomi et al., 2015). Renewable energy sources including solar, wind, and biomass energy (biohydrogen, biomethane, bioethanol, etc.) are expected to substitute the conventional energy sources i.e. fossil fuels in the near future (Hawkes et al., 2002). Among them, hydrogen has gained more attention because it is clean and environmentally friendly fuel, which

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produces only water rather than greenhouse gases when combusted (Bakonyi et al., 2013). In addition, the energy mass-based content of hydrogen is higher than other fuels as well as hydrogen can be produced from renewable substrates (Antonopoulou et al., 2008).

Several methods have been used to produce biohydrogen such as photo-fermentation, dark fermentation and biophotolysis of water (Antonopoulou et al., 2008). Among them, researchers have given more interest to dark fermentation because the process can be conducted in absence of light e.g. low energy consumption under ambient temperature and pressure as well as it can produce valuable products i.e. H_2, CH_4, etc., from waste substrates (Mohan et al., 2008). However, the main drawback of dark fermentation is the low hydrogen yield as maximum 33% of the electrons in the substrate can be converted into H_2, with 66% of the substrate electrons form soluble liquid metabolites (SLM) such as volatile fatty acids (VFAs), alcohol, etc. (Akinbomi et al., 2015; Bakonyi et al., 2018; Jung et al., 2012). In order to increase the total energy recovery and reduce the organic content, the H_2 fermented effluent should be further utilized in photo-fermentation, CH_4 production and microbial fuel cells (MFCs) (Jung et al., 2012).

In the recent years, two-stage fermentation for H_2 and CH_4 production has been proposed as a promising method to post-treat the H_2 fermented effluent and increase the total energy value of the process (Chu and Wang, 2017). During the two-stage process, hydrogen is produced in the first stage i.e. acidogenesis, then the liquid effluent of the H_2 fermentation is used as feedstock for CH_4 production in the second stage i.e. methanogenesis (Bakonyi et al., 2018; Luo et al., 2011). The effluent of the H_2 dark fermentation contains high volatile fatty acids (VFAs) concentrations including acetic acid, butyric acid, etc. (Algapani et al., 2018). Two-stage process for H_2 and CH_4 production has been widely investigated in previous works using various substrates and different operating conditions (Antonopoulou et al., 2008; Chu and Wang, 2017; Park et al., 2010).

The authors reported that the major obstacle of the H_2/CH_4 combined system is the low H_2 and CH_4 yields especially when agricultural residues are used as feedstock. This behavior may be due to the complex structure of the lignocellulosic matrix that can reduce the hydrolysis of the waste substrates (Monlau et al., 2015). Authors reported that application of pre-treatment methods such as acid, alkaline, hydrogen peroxide (H_2O_2), aerobic pre-treatment, etc., showed positive impact on the efficiency of the anaerobic digestion (Cheng et al., 2015; Monlau et al., 2015; Rafieenia et al., 2017). These pre-treatments are required to improve the biodegradability of the substrates and thus increase the H_2 and CH_4 yields (Pakarinen et al., 2009). For instance, Khatri et al. (2015) found that the methane yield increased from 302.81 mL-CH_4/g-VS using untreated maize straw to 313.35 and 370.99 mL-CH_4/g-VS when 4% and 6% NaOH pre-treated maize straw were used. Zhang et al. (2015) reported methane yield of 288 mL-CH_4/g-VS using 3% NaOH pre-treated rice straw compared to 187 mL-CH_4/g-VS for untreated rice straw. Cesaro and Belgiorno (2013) found that the biogas volume increased by 37% using ozonation pre-treated solid organic waste (0.16 g O_3/g TS ozone dosage) compared to the untreated substrate. However, few studies have considered the effect of pre-treatment on two-stage process for H_2 and CH_4 production so far and most of the studies have been conducted in batch assays. Therefore, more investigations are required to study the long-term H_2 and CH_4 production from pre-treated wastes. Such investigations are necessary to assess the long-term operation before application in industry and provide data about the energy and economical assessments (Monlau et al., 2015).

In a previous study, the effect of pre-treatment methods including thermal, alkaline, acid, H_2O_2, ultrasonication alone or in combination on biohydrogen production from potatoes and bean wastes was investigated in batch tests. Higher biohydrogen production was obtained using pre-treated wastes than the control tests. Thermal, ultrasonic pre-treatments did not show a great impact on the biohydrogen production, while alkaline, acid and H_2O_2 pre-treatments showed a significant influence on biohydrogen production. In addition, continuous biohydrogen production from organic wastes (sucrose, potatoes and bean wastes) was studied at different hydraulic retention time (HRT), of 24, 18 and 12 h. In case of potatoes and bean wastes, optimum hydrogen yields were observed at high HRT (Salem et al., 2018). However, it was observed that the removal efficiency of hydrogen fermentation was not high and the H_2 fermented effluent was rich in VFAs that could be used in a subsequent step.

Based on the findings of the previous study and the scarce of the research studies that considered the effect of pre-treatment on continuous hydrogen and methane production, the main objectives of the present study are: (a) investigating the effect of pre-treatment methods (alkaline, acid and H_2O_2) on bio-hydrogen and bio-methane production using potatoes and bean wastes, (b) analyzing the process parameters such as hydrogen and methane production (yield), VFAs concentration, etc., and (c) evaluation of the energy production efficiency in one-stage system for H_2 production and two-stage process for H_2 and CH_4 production using raw (or pre-treated) wastes.

2. Materials and methods

2.1. Seed sludge

The inoculum used in bio-hydrogen production experiments was obtained from Kasslerfeld wastewater treatment plant (Duisburg, Germany), and sieved through a mesh (2 mm) to remove waste materials. Thermal pre-treatment (at 100 °C for two hours) was applied on the bacterial species before inoculation to gather the hydrogen-producing bacteria such as *Clostridium species* as well as inhibit the activity of the hydrogen-consuming species e.g. methanogens and other non H_2-producing microorganisms. The heat-shock pre-treatment has been recognized as simple, effective and fast process compared to other inoculum pre-treatment methods such as chemical, acid/base, etc. (Bakonyi et al., 2014). For methane production, the sludge was collected from lab-scale continuous stirring tank reactor (CSTR). The CSTR was operated at HRT of 25 d for methane production from sewage sludge.

2.2. Feedstock preparation and characterization

The wastewaters were prepared as reported previously (Salem et al., 2018). Table 1 shows the characteristics of the feedstock substrates before and after the pre-treatment. The experimental results presented are the average measurements performed throughout the operation of the bioreactors. Three different pre-treatment methods as alkaline, acid and H_2O_2 were studied. The pre-treatment processes were performed as reported previously (Salem et al., 2018). The bioreactors were operated at feed and drain mode using peristaltic pumps. A nutritional solution was used as mentioned in previous work (Salem et al., 2017).

2.3. Experimental procedures

Two parallel CSTRs (active volume of 4 L) were used for biohydrogen production; the bioreactors were operated at HRT of 24 h and continuously mixed at 150 rpm. pH–control was used to maintain the pH at 5.5 by addition of 2 M NaOH and/or 4 M HCl according to the system requirements (iks Aquastar version 2.XX, iks Computer System GmbH, Germany). The feedstock tanks were continuously mixed to avoid sedimentation of the particles. The effluents from the H_2-producing bioreactors were collected in sedimentation tanks, and then were pumped into the CH_4-producing bioreactors. Methane was produced in up-flow anaerobic sludge blanket bioreactors (UASB). Two bioreactors: UASB-1, inner diameter 22 cm and height 140 cm, and UASB-2, inner diameter 24 cm and height 81 cm, with working volumes of 47.5 and 34 L, were operated at HRT of 13.6 and 12.1 d using H_2 fermented potatoes and bean wastes, respectively. The experiments were performed at mesophilic temperature of 35 °C by circulating water in the