



Gas biofuels from solid substrate hydrogenogenic–methanogenic fermentation of the organic fraction of municipal solid waste

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ABSTRACT

The objective of this work was to evaluate the performance of a two-stage hydrogenogenic–methanogenic (H–M) semi-continuous process in terms of mass retention time (MRT) for hydrogenogenic stage (H-stage), feed source for methanogenic stage (M-stage) and thermal regime (35 and 55 °C) for both stages. The substrate was a model organic fraction of municipal solid wastes (OFMSW) at 35% total solids.

In H-stage, mesophilic temperature had a positive significant effect on higher hydrogen productivities and lower amounts of hydrogen sinks compared to thermophilic operation. Calculations based on mass balances and biochemical stoichiometry confirmed that acid fermentation deviation was linked to low biohydrogen yields. The M-stage performance was influenced by both the temperature and feed source. Bioreactors in thermophilic regime performed better than mesophilic ones. Maximum methane productivity was 341 NmL CH₄/(kg_{wmr} d) that corresponded to the thermophilic bioreactor fed with fermented solids from H stage at 14 d MRT. The two-stage process showed higher gross energetic potential when compared to an only methanogenic process operated at equivalent MRT (control); this was due to a higher methane productivity in the M-stage of the series process. The main contribution of H-stage seemed to be associated to hydrolysis of the complex substrate thus generating metabolites for the M-stage rather than the hydrogen production itself.

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1. Introduction

In the last 15 years there has been an intensive research in order to improve and foster biohydrogen and methane technologies for biofuel production. From dark fermentation to cyanobacteria and purple non-sulphur bacteria, studies have focused on better hydrogen yields that can compete with H₂ costs from traditional non-renewable fuels [1]. As for biomethane, its exploitation has become an accepted/commercial technology [2–4].

Recently, interest on two-stage processes, hydrogenogenic followed by a methanogenic stage (H–M), has resurrected in the perspective of reclaiming bioenergy from organic wastes [5–9]. Two-stage process has been previously investigated in the way of an acidogenic coupled to a methanogenic bioreactor for waste sludge processing (two-phase anaerobic digestion), particularly during the seventies and eighties decades [10–12], aiming to enhance the methanogenic stage of sludge digestion. The main

objective of such an array was to separate the major microbial populations in different bioreactors according to its biochemical characteristics. Hydrolysis of the organic matter and its conversion to short-chain organic acids would occur in the acidogenic bioreactor, and so, microorganisms in the coupled methanogenic reactor would profit from this and further degrade the substrate and organic metabolites to methane and CO₂. Thus, methane yields have been improved when compared to one-phase process [13].

However, since hydrogen is produced along with organic acids such as acetate or butyrate, the opportunity to obtain biohydrogen as a fuel was then considered [2]. By means of the two-stage process it is possible, in principle, to attain a more thorough use of the substrate associated to the depletion of the organic load (as COD or VS), as well as increased yields of bioenergy and other valuable by-products [5,14–16].

So far, there has been a predominance of studies dealing with hydrogen and methane production using model substrates as sucrose, starch, or liquid wastes in submerged culture [17–19]. Yet, real application is not feasible for pure, expensive substrates. Therefore process success and scale up depends on processing inexpensive and almost inexhaustible substrates, such as organic wastes [20].

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Notations

A'	constant in Eq. (11)
A/B	acetic acid-to-butyric acid ratio on COD basis, dimensionless
B'	constant in Eq. (11)
C	constant in Eq. (8)
E_{CH_4}	gross energetic potential of M-stage (J) in Eq. (2)
E_{H_2}	gross energetic potential of H-stage (J) in Eq. (1)
E_{HM}	gross energetic potential of the H–M process (J) in Eq. (3)
\hat{E}_{HM}	specific gross energetic potential the H–M process (J/g _{db}) in Eq. (4)
H–M	two-stage (series) hydrogenogenic–methanogenic process
H-stage	hydrogenogenic stage (first stage of the H–M process)
I_{CH_4}	methane productivity (NmL CH ₄ /(kg _{wmr} d))
I_{H_2}	hydrogen productivity (NmL H ₂ /(kg _{wmr} d))
K_1	equilibrium constant of a given biochemical reaction at the lowest temperature T_1 in Eq. (7)
K_2	equilibrium constant of a given biochemical reaction at the highest temperature T_2 in Eq. (7)
k	specific rate of substrate uptake (g substrate/(g biomass d)) in Eq. (9)
k_d	endogenic decay coefficient (1/d) in Eq. (9)
K_s	affinity constant of substrate in Monod kinetic model (g/L) in Eq. (9)
LAB	lactic acid bacteria
$m_{i,H}$	mass inflow to H-stage (g _{wb} /d)
M	mass reactor content, (kg _{wb}) in Eq. (8)
m_f	mass flow of feed, (kg _{feed,wb} /d) in Eq. (8)
MRT	mass retention time
MRT _c	critical or wash-out retention time, that is, the retention time at which or below which the active biomass flows out of the reactor at a higher rate than its synthesis in Eq. (9)
MSE	mean of the sum of squares of the error
M-stage	methanogenic stage (second stage of the H–M process)
NL	volume in liters, normalized at 273 K and 101.325 kPa
NmL	volume in millilitres, normalized at 273 K and 101.325 kPa
OFMSW	organic fraction of municipal solid waste
Q_{CH_4}	methane production (NL/d)
Q_{H_2}	hydrogen production (NL/d)
r	number of replicates in the experimental design
R	ideal gas constant
S_0	concentration of substrate in the feed (g/L) in Eq. (9)
SEE	standard error of the experimental design
T	absolute temperature in Eq. (6)
T_1	lowest absolute temperature in Eq. (7)
T_2	highest absolute temperature in Eq. (7)
TS	total solids
TS _{i,H}	the total solids in hydrogenogenic stage inflow (%)
VOA	volatile organic acids excluding lactic acid
VS	volatile solids
[VS _f]	volatile solids content of the feed (kg VS/kg _{feed,wb}) in Eq. (8)
Y	biomass/substrate yield (kg biomass/kg COD removed), in Eq. (8)
Y_{CH_4}	methane yield (NmL CH ₄ /g VS _{removed})
Y_{H_2}	hydrogen yield (NmL H ₂ /g VS _{removed})

Greek characters

α	ratio between intermediate alkalinity to partial alkalinity in extracts from solid digestates, dimensionless
η	removal efficiency of VS, (kg VS _{removed} /kg VS _{fed}) in Eq. (8)
ρ	factor or ratio $\Sigma VOA/\Sigma Solvents$ on COD basis, dimensionless
$\Delta G_{T,P}^0$	change of standard free energy of Gibbs of a given biochemical reaction, at constant T and P (kJ/reaction) in Eq. (6)
ΔH_C	either hydrogen or methane combustion enthalpy (kJ/g) in Eq. (1) or Eq. (2)
ΔH^0	standard enthalpy change of a given biochemical reaction (kJ/reaction) in Eqs. (6) and (7)
ΔS^0	standard entropy change of a given biochemical reaction (kJ/K reaction) in Eq. (6)
$\Sigma Solvents$	total (sum) concentration of solvents, on COD basis
ΣVOA	total (sum) concentration of acetic, propionic, and butyric acids, on COD basis

Subindices

db	dry basis
ds	dry substrate
wb	wet basis
wmr	wet mass content of the reactor

The organic fraction of municipal solid waste (OFMSW) is a carbohydrate-rich waste considered as a potential biofuel producer [8,21,22]. Furthermore, organic solid waste is constantly produced by our society and it constitutes a major management and disposal problem for large cities all over the world, irrespective of developed or underdeveloped countries [23,24]. In Mexico, 102,000 ton/d are produced [25] and the organic fraction (OFMSW, paper and organic wastes) represents up to 60% of the total. Currently, efforts in some developed countries are being carried out on the evaluation of different technologies of waste reclaiming and disposal. Most important ones, other than dumping sites and landfilling, are fermentation/digestion, composting, and incineration [26], although increasing interest is being devoted to integrated approaches such as biorefinery of OFMSW [8].

It has to be considered that two-stage hydrogen-methane producing processes can be operated either on submerged cultures or on solid substrate fermentation mode. OFMSW applied to biofuel production has been mainly investigated on submerged or sludge fermentation modes [5,6,15,27,28]. Table 1 displays a comprehensive compilation of results of H–M processes published in the open literature. For instance, Lee et al. [28] reported the performance of a thermophilic two-stage fermentation process designed to produce H₂ and CH₄, using 10% TS food waste as feed. The system had also provisions for nitrogen removal and pH adjustment of sludge by returning the latter as an alkali buffer from the denitrification tank, hence leading to an actually H–M–DN process (DN stands for denitrification). Recirculation of high-alkalinity sludge (ca. 7.1 g/L as CaCO₃) seemed to be beneficial for long-term stability of the continuous process. Similarly, Chu et al. [29] produced hydrogen and methane from OFMSW (11.7% TS) on thermophilic regime with recirculation of sludge from methanogenic process. They asserted that the alkalinity and hydrogen-producing organisms still present in the recirculated sludges influenced positively the pH stability and hydrogen production. Although these references reported relatively long periods of stable operation of their processes, they are

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