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Dynamic one-dimensional model for biological methanation in a stirred tank reactor

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HIGHLIGHTS

- Modelling of hydrodynamics, gas–liquid mass transfer and biological reactions in a continuously stirred tank reactor.
- General model framework can be utilized for different reactor designs and biocatalysts.
- High gas–liquid mass transfer rate is the most critical parameter for high output gas quality.
- Scale-up study predicts stirring power to be 0.7–1.1% of the electrolyser power in order to reach over 98% CH_4 gas output.
- Dynamic simulations show fast response to inflow transients.

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ABSTRACT

Power-to-gas technology can facilitate the transition toward a renewables-based energy system by converting excess electricity to hydrogen and then into methane via methanation. Unlike traditional chemical methanation, biological methanation uses an aqueous solution of biomass (archaea), which consumes H_2 and CO_2 to produce CH_4. The process is limited primarily by the gas–liquid mass transfer step.

In addition to experimental research, modeling is often used to guide and expedite the development and scale-up of bioreactors from the laboratory to the pilot and commercial scales. Modeling has been used to optimize and test various operation conditions outside the range of experimentation. Estimations of gas–liquid mass transfer and the related stirring power are important for optimization and feasibility studies in the application of biological methanation to power-to-gas systems. Related published literature, however, is limited.

In this study, a dynamic model for a continuously stirred biomethanation reactor was developed with novel approach that combines semi-fundamental modeling of gas–liquid mass transfer, hydrodynamics, and biological reactions. The model was validated against existing experimental data and used in a sensitivity analysis of critical parameters, a scale-up study of a biomethanation reactor, and process dynamics studies. In each of the varying operational conditions, the model reproduced the trends observed in the experimental studies. The sensitivity analysis showed that biological parameters have a minimal effect on methane production. Conversely, the model is very sensitive to the gas–liquid mass transfer properties, such as the geometry of the impeller and reactor. The scaled-up study of biomethanation reactors with a CH_4 production capacity of 56–508 Nm^3/h revealed that the required stirring power is 0.7–1.1% from the electrolyzer power and decreases as the size of the reactor increases. High output quality (~98%) of the methane could be reached in each of the studied cases, and the overall efficiency of the power-to-methane process was roughly 50%. Dynamic simulations showed that the modeled process is tolerant to large gradients in the input parameters. After correctly setting the reactor- and process-specific parameters, the model can be used to perform scaled-up and dynamic studies of various reactor designs and different biomass solutions.

1. Introduction

Power-to-gas (PtG) technology can help mitigate the fluctuations caused by intermittent renewable electricity production by enabling the storage of excess electricity in the form of CO_2-neutral fuel. Although some pilot plants already operate using this technology, PtG is not used commercially [1]. Methane (CH_4) is a promising energy carrier option for PtG applications because the existing infrastructure for natural gas

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Nomenclature

A	cross-sectional area of the reactor [m^3]
a	interfacial area [m^{-1}]
b	constant [–]
c_g	gas concentration [$mol \cdot m^{-3}$]
c_l	concentration in liquid [$mol \cdot m^{-3}$]
c_X	Biomass concentration in liquid [$g \cdot L^{-1}$]
c^*_l	solubility in liquid [$mol \cdot m^{-3}$]
d_b	average bubble diameter [m]
d_{im}	impeller diameter [m]
d_{re}	reactor diameter [m]
D_L	liquid feed rate [s^{-1}]
D_g	gas diffusivity in liquid [$m^2 \cdot s^{-1}$]
g	gravitational constant [$kg \cdot m^{-1} \cdot s^{-2}$]
h	reactor height [m]
ΔH°	enthalpy of formation [$kJ \cdot (C)\text{-}mol^{-1}$]
H_g	Henry solubility [$mol \cdot m^{-3} \cdot bar^{-1}$]
I	inhibition factor due to the lack of substance [–]
k_D	saturation constant for H_2 [$mol \cdot m^3$]
k_L	mass transfer coefficient [$m \cdot s^{-1}$]
$k_L a$	volumetric mass transfer coefficient [s^{-1}]
m_X	maintenance constant [$(C)\text{-}mol_{H2} \cdot (C)\text{-}mol_X^{-1} \cdot s^{-1}$]
$\dot{m}$	mass flow rate [$kg \cdot s^{-1}$]
M	molar mass [$g \cdot mol^{-1}$]
N	stirring speed [rps]
n	number of nodes in grid [–]
n_{im}	number of impellers [–]
N_p	power number [–]
N_{is}	inter-stage number [–]
N_{ci}	circulation number [–]
p	pressure [Pa]
p_h	hydrostatic pressure [Pa]
p_{re}	reactor overhead pressure [Pa]
P_t	stirring power [W]
P_{ael}	electrolyser power [W]
Q	heat flux [W]
q^{max}	maximum specific H_2 conversion rate [$(C)\text{-}mol \cdot (C)\text{-}mol^{-1} \cdot s^{-1}$]
r	volumetric conversion rate [$(C)\text{-}mol \cdot m^{-3} \cdot s^{-1}$]
r_Q	heat dissipation [$W \cdot m^{-3}$]
S_g	source term for gas phase [$mol \cdot m^{-3} \cdot s^{-1}$]
S_l	source term for liquid phase [$mol \cdot m^{-3} \cdot s^{-1}$]
t	time [s]
T	temperature [°C]
u	velocity [$m \cdot s^{-1}$]
U	superficial velocity [$m \cdot s^{-1}$]
V	active reactor volume [m^3]
$\dot{V}$	volume flow rate [$m^3 \cdot s^{-1}$]
$\dot{V}_{L,is}$	inter-stage liquid flow rate [$m^3 \cdot s^{-1}$]
$\dot{V}_{L,ci}$	circulation liquid flow rate [$m^3 \cdot s^{-1}$]
y	component fraction is gas phase [–]
Y	(C)-molar yield of substrate [$(C)\text{-}mol \cdot (C)\text{-}mol^{-1}$]

Subscripts

ael	alkaline electrolyzer
C	CO_2
ci	circulation flow
D	H_2
G	gas phase
g	gaseous component
h	hydrostatic
is	inter-stage flow
L	liquid phase
l	absorbed gas component in liquid
im	impeller
N	NH_3
P	CH_4
re	reactor
tot	total
W	H_2O
X	biomass (archaea)
0	ungassed conditions

Greek

α	constant [–]
β	constant [–]
Γ	axial mixing coefficient [$m^2 \cdot s^{-1}$]
ε	energy dissipation [$W \cdot kg^{-1}$]
η	efficiency [–]
λ	liquid recycle constant [–]
μ	dynamic viscosity [Pa·s]
ρ	density [$kg \cdot m^{-3}$]
σ	surface tension [$N \cdot m^{-1}$]
ϕ	gas hold-up without viscous effects [–]
ϕ_υ	gas hold-up with viscous effects [–]

Abbreviations

CSTR	continuous stirred tank reactor
LHV	lower heating value
PtG	power-to-gas
vvd	gas volume flow per liquid volume in a day
vvm	gas volume flow per liquid volume in a minute

transportation can be used and the methanation reaction can use the CO_2 emissions as a raw material. There are several possible routes for methanation that can roughly be divided into chemical and biological types [2]. The main advantages of biological methanation are simpler equipment, low operational temperature (< 70 °C), higher tolerance for impurities in the input gases, and less complicated dynamic operation than those of the corresponding chemical process [1]. Moreover, biological methanation offers the possibility to increase carbon yield from municipal, agricultural and food waste—the key carbon sources in urban areas—aiming for 100% renewable and self-sufficient energy production. Dynamic modeling tools are essential for the intelligent control and profitable operation of future energy systems, where materials of highly variable composition, such as wastes, are utilized in conjunction with the generation of renewable electricity.

A broad review of biological methanation was done by Rittman et al. [3]. They noted that the gas-liquid mass transfer is often the limiting step of the process, and that the stirring power should be minimized, while maintaining a high conversion efficiency. However, no power consumption metrics were published in the reviewed literature. The gas-liquid mass transfer and scale-up processes for bioreactors were extensively reviewed by Garcia-Ochoa and Gomez [4], underlining the complexity in scaling up. A comparison of several reactor designs was performed by Kougias et al. [5], who found that a double bubble column had the best performance. Thus far, techno-economic and life cycle analyses have only been conducted for PtG-systems with traditional chemical methanation, as in recent studies by Parra et al. [6], Collet et al. [7] and Zhang et al. [8].

Several experimental studies on biological methanation have been performed at the laboratory scale with biofilm plug-flow [9], fixed bed [10], trickle-bed [11–13], closed batch [14] and continuously stirred