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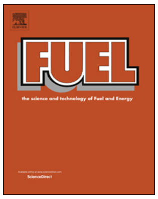

Review article

Overview of recent progress towards in-situ biogas upgradation techniques

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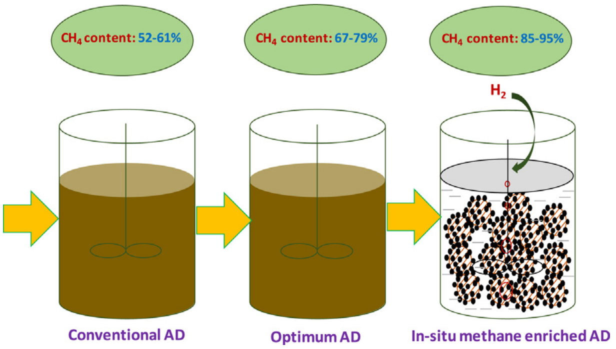
GRAPHICAL ABSTRACT



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ABSTRACT

Biogas, as derived from the anaerobic digestion process, offers a versatile possibility of renewable and sustainable energy usage. When enriched, upgraded biogas can yield high levels of biomethane, allowing its use as an alternative to natural gas via existing natural gas grids or being directly consumed by transport vehicles as fuel. Currently, biogas upgrading is experiencing a golden period of rapid development where many enrichment techniques are being revisited, modified or strengthened, and contemporary novel technologies are being proposed. Mainly, two broad categories of upgrading techniques are present in which conventional method primarily focuses on *ex-situ* approaches, treating produced biogas to methane by employing catalytic conversion (biological and chemical), membrane gas-permeation, desulphurization, physical and chemical scrubbing, absorption and adsorption. Over the years, a considerable effort has been made to improve efficiency and to enhance the economic viability of the above techniques and many commercial plants worldwide use *ex-situ* approaches as options to enrich biogas as biofuel for direct utilization to vehicles. Coupled with the *ex-situ* method, *in-situ* techniques, such as CO_2 desorption, pressurized reactor, H_2 addition (deployed to anaerobic digesters directly) and electromethanogenesis has also been gained significant attention recently. Comparative studies between *in-situ* and *ex-situ* method suggest that the former provides an increased economic performance for small to medium and small-scale facilities, allowing the upgrading of biogas above 85% v/v of methane. Additionally, innovations in bacterial species that are capable of direct exchange of electrons, escalating the biological conversion of CO_2 to CH_4 has also been demonstrated. This paper enlightens some of these aspects and reviews the state-of-the-art of biogas enriching techniques emphasizing *in-situ* approaches.

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Nomenclature

AD	Anaerobic digestion
AHPD	Autogenerative high pressure digestion
BES	Bioelectrochemical system
CHP	Combined heat and power
DIET	Direct interspecies electron transfer
FA	Free ammonia
GHG	Greenhouse gas
GW	Giga watt
HRT	Hydraulic retention time
L/G	Liquid-to-gas
MEC	Microbial electrolysis cells
MFC	Microbial fuel cells
OLR	Organic loading rate
SHE	Standard hydrogen electrode
UASB	Upflow anaerobic sludge blanket
VFA	Volatile fatty acids

1. Introduction

As the availability of fossil fuels is constantly decreasing, there is an increasing concern towards reducing energy usage and production derived carbon dioxide emissions [1]. As a consequence, the demand for accelerating the growth of alternative energy sources has gained more public attention than ever before [2]. Wind, solar and biomass are the three main sources of renewable energy expected to cover the bulk of the future energy supply worldwide, replacing fossil fuels [3]. Many energy policies already reflect this shift and target a substantial volume of alternative energy in their future energy mix based on available resources and complying with the Kyoto protocol [4]. Unlike wind and solar energy technologies (which are termed as intermittent renewable energy technologies), biomass is abundant, versatile, and has a continuous power generation capability (once reliable logistics are guaranteed) [5], and currently accounts for 10% of primary energy supply worldwide [6].

There can be different routes of biomass conversion technologies [7]. Biochemical conversion that produces biogas using a variety of wastes and organic sources in a controlled anaerobic digestion process is suitable to fulfill part of the future sustainable energy production objective, as this method (when compared to thermochemical and thermal conversion techniques) is highly economical and efficient [8]. Wastes like animal manure, sewage sludge, municipal solids and agricultural residues are specifically important in the context of biogas because they do not compete with agricultural food crops [9]. On a global scale, the amount of anaerobically digested substrate increases remarkably with an annual growth rate of ~25% [10]. Biogas production, therefore, has potential to generate a large amount of energy. The elevation of anaerobic digestion capacity to allow increased waste treatment and biogas production have been emphasized a great deal in previous studies [11–14]. Currently, the installed electricity production capacity of anaerobic digestion plants within the European Union has reached close to 7.9 GW which in addition to heat production may rise close to 29.5 GW by 2022 [15].

Nevertheless, biogas is not readily suited to all energy applications, primarily because of its low level of heating value (calorific value) and impurities. Currently, the majority of the commercial biogas plants are operated as combined heat and power (CHP) where biogas fueled engines produce the required heat and electricity to meet the energy demands on site and to the external consumers [16]. However, since the electrical efficiency of commercial gas engines is low (between 30 and 40%), electricity produced from biogas based CHP is not competitive in the free electricity market without substantial government subsidies. An alternative route, as developed over the last few decades, is the upgrading of biogas to a higher level of methane quantity. This can be used either as compressed biomethane locally or as renewable fuel directly injected into the natural gas grid. The positive economic and energetic effect for substitution of fossil fuels with enriched biomethane from biogas instead of electricity derived directly from CHP has already been demonstrated [15] with the commercial interest growing continually.

In order to increase the methane content in biogas, especially for use as a transport fuel, a large number of innovative technologies have been developed [17,18] and recently reviewed [19–22]. The technological focus has generally been towards extensive cleaning and downstream processing of biogas by deploying techniques such as drying, and the removal of CO_2, NH_3, H_2S and other trace impurities to achieve a methane content of 95–99% in biogas. However, impurity removal can be of cost and energy intensive including technical barriers associated with low sorbent efficiency (sorbents or chemicals: i.e, alkaline amine, zeolites and metal–organic frameworks) [15] and plasticizing of membranes [23]. Past studies [24,25] have suggested that due the large fraction of CO_2 in raw biogas, the cost of gas purification only becomes economically and energetically feasible if plant operational capacity exceeds 100 m^3 biogas/h. A large number of real applications, however, operate below this range and thereby the development of *ex-situ* technique up until now is underemphasized. Today, only a very few commercial plants upgrade biogas to a high fuel standard using *ex-situ* cleaning of the biogas globally [26].

Through the *in-situ* technique, when applied to the anaerobic process directly operating with the concept of CO_2 and CH_4 differential solubility and electro-methanogenesis, a cost-effective way of upgrading methane over a broad range of applications may become established. To date, a number of methods regarding *in-situ* methane enrichment have been proposed and interesting results were demonstrated [26–31]. Besides being cost-effective, *in-situ* upgrading is deemed to offer enhanced degradation of organic matter [30] with simultaneous removal of H_2S from the off-gas (which is technically as expensive as removing CO_2 from biogas) [26]. Furthermore, in a novel electro-methanogenesis concept, several groups of bacteria can efficiently exchange electrons, directly producing biogas with high methane. Despite this, the research and development towards upscaling of various *in-situ* techniques are still ongoing.

Biogas upgrading using combined *ex-situ* and *in-situ* techniques have been reviewed by some published documents previously [12,15]. However, literature review reporting *in-situ* biogas upgrading only is scarce, if not none. The aim of this review, therefore, is to define the state-of-the-art *in-situ* biogas upgrading techniques and to shed light on innovations that could be employed for future advancement in biogas production technologies. In particular, the work explores various methodologies with emphasis on emerging processes, which are envisaged to play a significant role in the future context of bioenergy.

2. Biomethane enrichment

Raw biogas produced by the anaerobic digestion generally consists of the gas species CH_4, CO_2, H_2S, NH_3 and H_2O, along with the trace amount of other organic and inorganic components. Methane has a large share within the biogas composition with 40–75%, followed by CO_2 with 25–55% [12]. Besides anaerobic digestion, biogas can also be collected from landfills with a typical gas composition of 50–55% CH_4, 37–45% CO_2 and < 1% non-methane organic and inorganic compounds [12,32]. Regardless of the production routes, compared to its closest counterpart natural gas (fossil fuel), biogas is energetically inferior due to the high amount of CO_2 and other contaminants. Moreover, the lower heating value of biogas for example is roughly 21.5 MJ/Nm^3, while it is around 35.8 MJ/Nm^3 for natural gas [33].