

Carbon Capture and Utilization Technology Development Opportunities Based on Biomethanation

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Abstract

The paper provides an overview of Power-to-Gas (P2G) technology using biomethanation and a proprietary biocatalyst. It addresses the issue of carbon dioxide (CO₂) emissions from fossil fuel combustion and proposes the integration of Carbon Capture Utilization and Storage (CCUS) technologies with P2G processes. Currently, the integration of CCUS and P2G is in conceptual stage. The paper emphasizes the sensitivity of biocatalysts to contamination in feed gases, particularly the negative impact of oxygen on methanation processes. Findings from measurements conducted in 2022 using a lab-scale prototype approve that post-combustion technologies can be successfully integrated into P2G technologies through the utilization of biomethanation processes. Various parameters, such as Carbon Dioxide Conversion (CDC), Volumetric Methane Production (VVD), and Higher Heating Value (HHV), were calculated based on the measured datasets. The high CDC value of 96.65%_(v/v) and 68.03%_(v/v) of methane content indicates successful integration of the two technologies, while increasing the CO₂ source and applying higher pressure in the biomethanation reactor can further enhance VVD. In conclusion, the paper highlights the potential of P2G technology based on biomethanation and its integration with CCUS processes. The results obtained from the lab-scale prototype demonstrate promising conversion rates and suggest avenues for improving VVD.

Keywords

power-to-gas, biomethanation, decarbonization, CCUS

1 Introduction

The European Union focusing on finding sustainable solutions to address environmental, social and economic issues caused by climate change. One key pillar of the European Green Deal strategy [1], which aims to develop transition to build more integrated networks and support the use of renewable energy sources. Decarbonization of the gas sector, promoting innovative technologies and modern infrastructures are emphasized as crucial factors for the success of this transition. The 2009/28/EC the "RED I" [2] and 2018/2001/EU (RED II) [3] directives set targets for renewable share of total energy consumption in the EU increasing the target from 20% to 32% by 2020. The EU goals specified in these directives were transposed into the domestic legal frameworks, such as "National Energy Strategy 2030, with a view to 2040" [4] and "Hungary's National Hydrogen Strategy" [5]. In addition, the targets set in REPowerEU strategy, European Union needs to boost biomethane production to 35 bcm by 2030 [6, 7].

One of the opportunities for reaching climate neutrality and developing a state-of-art energy structure can be achieved by innovative biogas technologies, as well as technologies enabling seasonal energy storage, which can be integrated into an innovative Power-to-Gas (P2G) technology [8–11]. The draft directive, published in late December 2021, outlines the common rules for the biomethane and hydrogen market, specifically concerning their transportation through the natural gas network [12]. It forecasts the share of biomethane and hydrogen in the gaseous fuel market mix to be 66%, which prioritizes the use of P2G technologies using biomethanation processes. In line with the international biomethanation projects [13], the scientific and industrial R&D results realized achieved in the domestic P2G technology development [14–16] serve as a foundation for harnessing the potential in P2G technologies in Hungary as well, and speed up the transition towards the hydrogen economy [17]. The importance of P2G technologies

is further amplified due to the demand for grid balancing caused by photovoltaic power generation [18].

Experts of various fields of R&D related to energy generation, distribution and storage are making actively working to develop newer and more efficient technologies [19, 20]. Technological innovations target project ideas that facilitate the adaptation of the Hungarian energy sector to adapt to the changing trends (such as decarbonization or circular economy [21, 22]. Furthermore, the constantly growing body of international literature providing valuable insights, data, and research findings in the field, the utilization of novel material compositions enable the investigation of the technical and economic possibilities of biogas and biomethane production through P2G technology [23, 24].

Biomethanation using selected or mixed archaea/ bacteria culture offers a pathway to implement P2G technology that can convert raw biogas into valuable fuel without the need to separate its carbon dioxide (CO_2) component [25]. The P2G processes that convert the CO_2 component of biogas by using thermo-catalysts (e.g., nickel, ruthenium) are considered as alternative way to produce biomethane, which meets natural gas standards. The thermo-catalytic reaction is based on Sabatier process, which is carried out at temperatures of 250–400 °C with the pressure of 1–80 bar [26]. Volatile nature of RES can be addressed through P2G processes using a biocatalyst. A commercially-scaled P2G biomethanation plant has been operating in Avedore (DK) since 2016 [27].

The integration of Carbon Capture Utilization and Storage (CCUS) technologies with P2G technologies holds potential. During the combustion of fossil fuels, CO_2 is generated, and its separation (absorption) and utilization draws significant attention in line with the goal of climate neutrality. One of the primary types of CCUS strategies is post-combustion capture [28], which is a particularly suitable choice in the case of existing power plants since the post-combustion separation equipment after firing can be retrofitted [29, 30].

Considering the application of P2G at industrial sites, research on the integrating CCU processes with P2G systems is currently at conceptual stage. In an integrated concept, the initial step involves green hydrogen generation through electrolysis using renewable electricity. Subsequently, this green hydrogen can be further converted into carbon-neutral synthetic natural gas (SNG) through a biological methanation process known as power-to-methane (P2M). In this concept, CCUS plays a crucial role by capturing the CO_2 content of the flue gas from

the gas engine, providing the required CO_2 for the biological methanation process as illustrated in Fig. 1.

2 Material and methods

2.1 Technological background

2.1.1 Overview of carbon capture technologies

To achieve enhanced decarbonization and supply CO_2 for P2G processes, fossil-fuel plants can be equipped with various carbon capture technologies. Currently, three technological strategies are available: pre-combustion, oxyfuel combustion, post-combustion technologies [31].

1. *Post-combustion* solutions are considered the most mature and the most practical option for existing plants. Various technologies are available for post-combustion carbon capture technologies, including chemical absorption; physical absorption; adsorption; gas-particle reactions; membrane separation and cryogenic separation [32].
2. The *pre-combustion process* involves partial combustion of the fuel, resulting in the production of CO_2/CO and H_2 through processes such as partial oxidation. The H_2 is separated then from CO_2 using physical or chemical methods and used as a fuel, ultimately generating water as a combustion product. Although pre-combustion CC is highly effective in reducing pollution, it typically incurs higher overall cost and it has a lower overall performance [33]. The most common pre-combustion separation technique is physical sorbent-based separation [34].
3. While considered the least mature technology, *oxyfuel combustion* models are also a viable option [35]. In an oxyfuel combustion power plant, the fuel is oxidized in a nitrogen-free environment, resulting in combustion products primarily composed of CO_2 and water vapor. A relatively straightforward condensation and

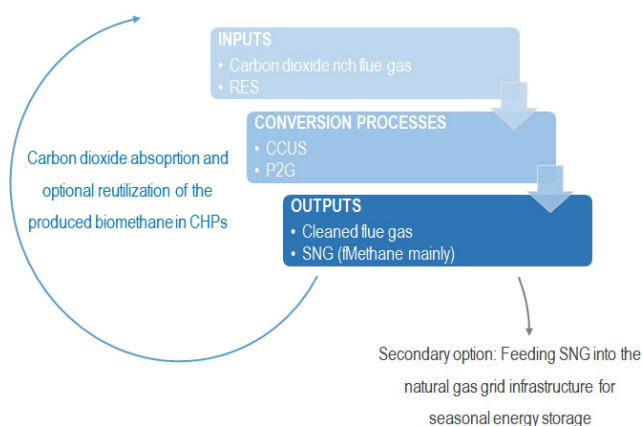


Fig. 1 Concept of P2G and CCUS technology integration

separation process enables cost-effective carbon capture and separation. However, implementing oxyfuel combustion requires new plant developments or modifications to existing facilities due to the higher operating temperatures involved [35].

Additionally, Direct Air Capture (DAC), a relatively new technology in its early commercial stages, offers an alternative for capturing CO₂ directly from the atmosphere or from diluted gases and distributed sources of carbon through industrial processes. However, it is important to note that this innovative approach has not yet been implemented on a commercial scale [36].

The most mature among the aforementioned processes is the post-combustion method, typically involving three main steps:

1. The flue gas emitted from a fossil fuel power plant is either vented directly into the air or directly into a solvent absorber;
2. The absorber contains a solvent that absorbs CO₂ from the flue gas. The solvent containing CO₂ is transported to a stripper for CO₂ regeneration;
3. In the stripper, the CO₂ is separated from the solvent. The separated CO₂ is compressed using compressors and then transported (through pipelines, ships or trucks) to suitable storage facilities such as depleted oil and gas reservoirs, unsinkable coal beds, saline aquifers and basalts [31].

Absorption technologies used in CC can be classified into two groups: physical absorption and chemical absorption. Depending on the type of absorbent employed, the absorption processes encompass various techniques, such as alkanolamines absorption, aqueous ammonia absorption, dual-alkali absorption, sodium carbonate (Na₂CO₃) slurry absorption, and the chilled ammonia process [37]. Based on the literature, the chemical absorption is considered the preferred method for capturing CO₂ from gas streams and low to moderate partial pressures of CO₂ (3–20%). It is regarded as the most developed method [38] and the only one that does not need further improvements before large scale implementation. Chemical absorption is frequently proposed in the literature as the preferred method for CO₂ capture in gas-fired plants [39]. The method's predominance is widely supported by several research groups (including mineralization or bio-fixation 21%; adsorption 14%; membranes 8%) [37]. Based on recent research and development results, post-combustion

capture utilizing chemical absorption has reached a mature stage as a technology. It holds the potential for large-scale deployment in power generation and in other energy-intensive industries relying on fossil fuels [40].

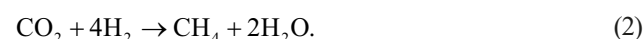
2.1.2 Power-to-Gas with biological methanation

Power-to-Hydrogen (P2H) and Power-to-Methane (P2M) are the two dominant segments of P2G. The P2H process involves water electrolysis, a widely-recognized technique for converting water into molecular hydrogen and oxygen using relatively low temperatures. This process also can be powered by renewable electricity. The reaction can be described as follows [41, 42]:



Currently, there are four different methods are used and/or studied for electrolysis: Alkaline-based (AEL), which is the most mature technology; Proton exchange membrane (PEMEL), which is also mature and offers more flexibility; Solid oxide electrolyzers (SOEL), which is highly efficient but still in the early stages of development; Anion exchange membrane (AEM), which combines the advantages of AEL and PEMEL. Additionally, small-scale units for AEM electrolysis are commercially available [43, 44].

In the second step of the process chain, CH₄ is formed by combining H₂ with CO₂. The necessary CO₂ can be sourced from several different outlets including biomass plants, power plants, and industrial processes or even extracted from ambient air. The methanation step plays a crucial role in converting H₂ and CO₂ to CH₄ and H₂O. [38] At this stage, there are two approaches, namely chemical and biological methanation, are seen as "competing" with each other. The reaction equation of the power-to-methane process can be written as follows [41, 42]:



Currently, there are four main types of reactors to can be used for the chemical methanation process: fixed bed, fluidized bed, three-phase, structured. Usually, continuous stirred tank reactors (CSTRs) are used for biological methanation. In CSTRs, the reaction takes place in an aqueous solution that is continuously agitated mechanically, which is essential for promoting efficient gas–liquid mass transfer [45].

In biological methanation, the reaction is catalyzed by one or multiple strains of microorganisms. To facilitate their growth and activity, the strains require suitable temperature and pressure conditions, as well as necessary

nutrients. These nutrients are typically provided as a solution to the reactor where the methanation takes place [46]. These microorganisms belong to a highly diverse group of archaea known as methanogens. Methanogens are characterized by their unique ability to produce methane gas (CH_4) through metabolic processes. They utilize CO_2 and hydrogen (H_2) gases, as well as small organic molecules such as acetate, formate, and methylamine, as substrates for methane production [47].

In contrast to catalytic conversion plants that have been operational for several years, the implementation of biological methanation has only recently reached industrial pilot scale and is approaching near-term commercial application [48]. Although biological methanation may have a lower reaction rate compared to other methods, there are multiple advantages that make it an attractive option:

- Biological methanation operates at relatively mild temperatures (between 35 °C and 70 °C) and pressures (between 1 and 10 bars) making it easier to employ and handle compared to high-temperature and high-pressure processes;
- Biological methanation is less sensitive to impurities in the feedstock compared to catalytic processes. Compounds such as H_2S and NH_3 can serve as nutrients for microorganisms involved in the process;
- In biological methanation, catalyst renewal is carried out continuously through microbial growth;
- Due to the milder operating conditions, reduced sensitivity to impurities, and continuous catalyst renewal, biological methanation generally has lower operating costs compared to other methane production methods [49].

Pure cultures can be characterized by the name of their species, strain and origin of culture. While most studies on biological methanation have focused on mixed cultures, there have been a few studies conducted using pure cultures such as *Methanobacterium*-like strain IM1 [50]; *Methanothermobacter thermoautotrophicus* IM5 [51].

Pure cultures are ideal for research projects since their methane productivity is higher and process conditions can be specified more precisely compared to a biocatalyst using mixed cultures. In addition, working with pure cultures reduces the risk of contamination by other microorganisms in the process and lower costs associated with medium preparation. Working with pure cultures may also present challenges. During the start-up operation, a longer inertization period might be necessary, as pure

cultures are more sensitive to oxygen exposure [51]. While mixed culture-based studies provide valuable insights into the specific characteristics of archaea strains, further researches on pure cultures are crucial to obtain precise data on aspects such as electroactivity [47].

Since the commercial application of the biomethanation technology is still at the early stages, a limited number of large-scale biomethanation projects have been undertaken so far (e.g., MicrobEnergy's large scale tests in both ex-situ and in-situ bioreactors; Krajete's laboratory scale tests; Electrochaeta GmbH's projects). Nevertheless, there are several researchers actively engaged in exploring novel approaches to enhance and improve biomethanation technology. For example, Therma et al. [51] conducted a study where they developed a fully automated process converting H_2 and CO_2 into methane using a high temperature trickle-bed reactor; Szuhaj et al. [52] tested the performance of a mixed culture in lab-scale power-to-methane reactors at 55 °C using a fed-batch system which upgraded the gas mixture to 95% bio CH_4 purity (close to the methane content required for direct injection into the natural gas grid) and produced a significantly higher yield than reported in previous studies; and Zipperle et al. [53] converted synthetic carbon monoxide (CO)-containing flue gases to methane (CH_4) by artificial hyperthermophilic archaeal co-cultures, consisting of *Thermococcus onnurineus* and *Methanocaldococcus jannaschii*, *Methanocaldococcus vulcanius*, or *Methanocaldococcus villosus* resulting in up to 10 %_(M/M) CH_4 .

2.2. The focal technology

Since April 2018, Power-to Gas Hungary Kft. has been operating an innovative laboratory-scaled P2G prototype that utilizes biomethanation process. The owned lab-scaled P2G prototype served as a valuable tool for conducting biomethanation research activities shown in Fig. 2.

The main components of the prototype are

- Polymer electrolyte membrane (PEM) electrolyzer producing hydrogen;
- CO_2 /mixed gas flow regulators;
- Water-jacketed continuous stirred tank reactor with 2Litre capacity and its operating control unit;
- Industrial gas analyzer.

The system incorporates flow regulators that enables precise control of the volume flows and the concentration of the H_2 : CO_2 /experimental gas mixture. The control unit regulates the doses of nutrient solutions required

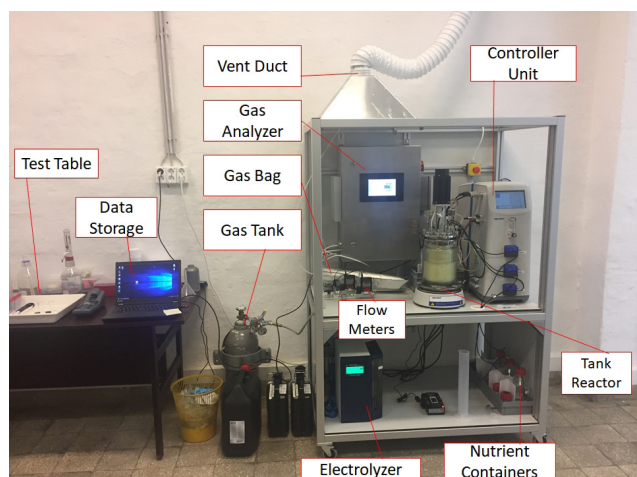


Fig. 2 Benchtop lab-scale prototype at the site of Power-to-Gas Hungary Kft.

for the optimal functioning of the biocatalyst. It also controls the speed of the mixing shaft and the desired temperature within the reactor. To monitor the progress of the biomethanation process, a gas analyzer employed. At pre-programmed intervals, it measures the concentration of methane, H_2 and CO_2 in the off-gas. Eppendorf BioFlo 120 controller is used for feeding essential nutrients such as ammonium hydroxide solution (NH_4OH , 4.5% (2.41M) and sodium sulfide solution (Na_2S , 0.042M (5.5g/L)) into the reactor. Ammonium hydroxide serves as the primary nitrogen source for microorganisms, while the sodium sulfide solution supports the growth of the culture by providing necessary sulfur.

Onsite measurements play crucial role in evaluating the required volume of the two media and monitoring the health of the living culture. After measuring the volume of the discharged substrate, several essential parameters are monitored to assess the health and performance of the biomethanation process, including optical density, oxidation/reduction potential and pH level.

The biocatalyst contains microorganisms that absorb nutrients in liquid, dissolved form for methane synthesis. At the same time, compliance with the main physical parameters for the biocatalyst is a prerequisite for methanation. These parameters typically include a 60–70 °C, 7.5–8.5 pH, $CO_2 \leq 0.5\%_{(V/V)}$. Market demands justify, that the fully mature solution that satisfies all these criteria and has reached a technology readiness level (TRL) above 8 is currently not available [21].

2.3 Measurement description

The prototype unit produces ca. 3.6 l/h methane at regular setups. The prototype unit produces ca. 3.6 l/h methane

at regular setups. Normal operating experiments are carried out in campaigns of 100 or 200 hours. To ensure the health of the single-cell biocatalyst (*Methanothermobacter thermautotrophicus*), a systematic start up and shut down protocols have been followed. During operation, the CO_2 dosage has been carefully adjusted according to specific requirements. At times, the operation may be temporarily halted, allowing for the removal of approximately 100–200 mL of metabolic water. This removal process typically takes place each day between 7.00 and 7.30 a.m. To support the growth and vitality of the biocatalyst, solutions containing essential micronutrients (such as Mn, Co, Cl, Ni, Se, W) are injected into the tank reactor through the top septum.

To ensure the health of the living culture, regular measurements are conducted as part of the monitoring process. Based on the volume of the discharged substrate and the conducted OD, ORP and pH level measurements, the required volume of the concentrated media can be evaluated. The results of the tests are systematically recorded in a table including the following specifications:

- Date,
- pH value,
- Stirring rpm (min^{-1}),
- ORP (V),
- OD (%),
- H_2 flow rate ($mL min^{-1}$),
- CO_2 flow rate ($mL min^{-1}$),
- CH_4 composition ($\%_{(V/V)}$),
- H_2 composition ($\%_{(V/V)}$),
- CO_2 composition ($\%_{(V/V)}$),
- Nitrogen source pump flow rate ($mL min^{-1}$),
- Sulfide pump flow rate ($mL min^{-1}$),
- Comments.

Carbon Dioxide Conversion (CDC) and Volumetric Methane Production (VVD) rates are calculated using Eqs. (3) and (4):

$$CDC = \frac{c_{\text{methane}}}{(c_{\text{methane}} + c_{\text{carbon dioxide}})} \times 100[\%], \quad (3)$$

$$VVD = \frac{CC}{2L \times 100} \times V_{CO_2} \times 60 \frac{\text{min}}{h} \times 24 \frac{h}{\text{day}} \left[\frac{L}{L \times \text{day}} \right], \quad (4)$$

where:

- c_{methane} and $c_{\text{carbon dioxide}}$ stand for methane and CO_2 concentration in off-gas ($\%_{(V/V)}$);
- V_{CO_2} is volumetric flow rate of CO_2 measured in $sL min^{-1}$, where sL is 1 liter volume of the gas mea-

sured at standard sets of conditions ($t = 0\text{ }^{\circ}\text{C}$, $p = 101.325\text{ kPa}$).

3 Results and discussion

The operating campaign chosen for the analysis was conducted between July 25 and 29, 2022. During this period, specific gas and nutrient pump flow rates were adjusted and utilized. These flow rates are presented in Table 1, providing a clear overview of the applied settings for the gas and nutrient supply during the campaign.

In the 100-hour-campaign, measurements were recorded at a 20-minute interval, resulting in the collection of over 850 records regarding the off-gas composition. These records include measurements related to air composition as well as sensor health data. To ensure the accuracy of the analysis, the air measurements and sensor health data are carefully filtered and deleted. Once the data is processed, CDC and VVD are calculated using Eq. (3) and Eq. (4). The graphs displaying the measured concentrations ($\%_{(V/V)}$), CDC (%) and VVD ($\text{L L}^{-1}\text{ day}^{-1}$) are shown in Fig. 3.

The process of metabolic water removal and the subsequent OD, pH, and ORP measurements required a total of 15 minutes. Following the operation protocols of the

prototype, it was necessary to shut down the gas flow regulators, gas analyzer, and agitator during this period. Due to temporary halt in off-gas production and the relatively small scale of the reactor tank, there were periodic drop in the methane production on daily basis. This decrease in methane production was accompanied by the increase of H_2 and CO_2 concentrations. Nevertheless, sudden changes in the off-gas composition unlikely occur since the excess metabolic water is discharged continuously into the sewage grid infrastructure. Table 2 presents the detailed results.

At the start of the campaign during the ramp-up phase of the biochemical activity of the biocatalyst, the minimum methane concentration was observed while the maximum value of CO_2 and H_2 off-gas component concentrations were registered. These initial conditions contributed to the lower values of CDC and VVD. To enhance the methane concentration, the use of a pressurized tank reactor could be beneficial. This could potentially increase the average methane concentration, which was recorded at $68.03\%_{(V/V)}$ during the campaign. The average carbon dioxide concentration during the campaign was recorded

Table 1 Gas flow regulator and nutrient pump setup values

Name	Value
CO_2 (sL min^{-1})	0.06
H_2 (sL min^{-1})	0.252
NH_4OH pump H_2 (mL min^{-1})	0.03
Na_2S pump (mL min^{-1})	0.04

Table 2 Maximum, minimum, mean and standard deviation (SD) of off-gas component concentration datasets, CDC and VVD calculations

Name	Max	Min	Mean	SD
CH_4 concentration ($\%_{(V/V)}$)	75.78	1.81	68.03	11.1
CO_2 concentration ($\%_{(V/V)}$)	8.43	0.01	1.32	1.5
H_2 concentration ($\%_{(V/V)}$)	87.38	13.6	25.91	8.33
CDC (%)	99.99	19.9	96.65	7.95
VVD	43.2	8.6	41.35	3.44

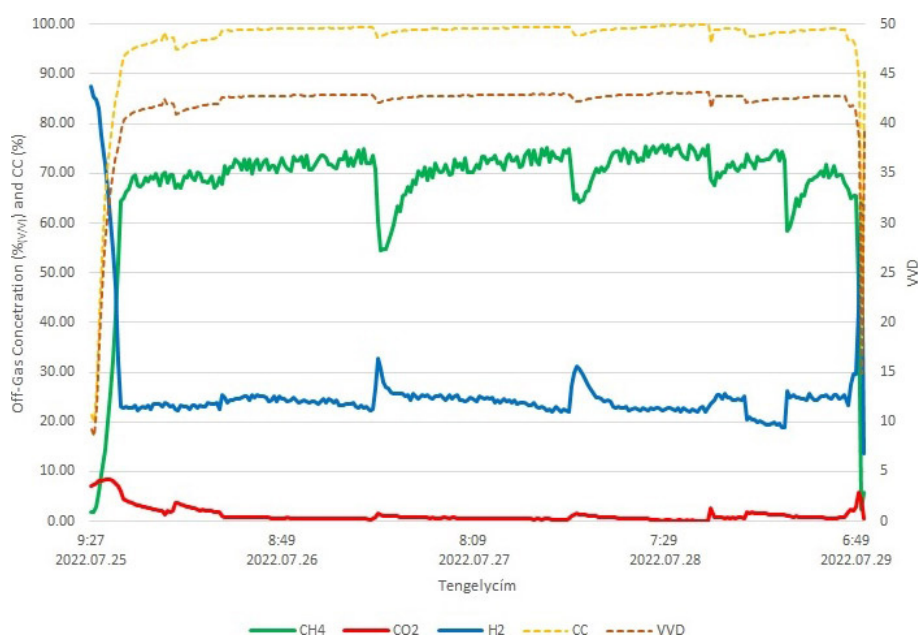


Fig. 3 Off-gas composition measurements, CDC and VVD calculations

at 1.32%_(v/v). To comply with effective natural gas market standards of the EU, reduction in carbon dioxide concentration is required [54].

The VVD mean (41.34 L L⁻¹ day⁻¹) was limited by the volume of the gas bag, buffer storage of the off-gas, which is led into the gas analyzer. Despite this limitation, the VVD value is considered adequate comparing to other lab-scale experiments [55].

Measurements conducted to assess the health of the biocatalyst showed only minor variations throughout the campaign. The value of pH 8.2–8.3; ORP (–400)–(–420 mV) and OD 35–40% confirmed the eligibility of the measurement conditions.

The CDC rate achieved during the campaign was extremely high, with the median (2nd quartile) of almost 99% (98.98%). This indicates the high selectivity of the biocatalyst in converting carbon dioxide into methane. To determine the Higher Heating Value (HHV) of the off-gas mixture, the off-gas concentration of each component is multiplied by its respective GCV and the sums of these products are added together as shown by Eq. (5):

$$\text{HHV}_{\text{off-gas}} = c_{\text{methane}} \times \text{HHV}_{\text{methane}} + c_{\text{carbon dioxide}} \times 0 + c_{\text{hydrogen}} \times \text{HHV}_{\text{hydrogen}}, \quad (5)$$

where:

- $\text{HHV}_{\text{methane}} = 35.8 \text{ MJ/sm}^3$;
- $\text{HHV}_{\text{hydrogen}} = 12.71 \text{ MJ/sm}^3$;

Using mean values of Table 2 and Eq. (5), the calculated GCV for the produced off-gas is 24.64 MJ m⁻³.

4 Conclusion

The measurement results obtained from the lab-scale prototype demonstrate the suitability of *Methanothermobacter thermautotrophicus* archaea biocatalyst for CCUS integrated biomethanation application. The average off-gas CO₂ component concentration was 1.32 %_(v/v) indicating a significant reduction in CO₂ content through the biomethanation process. The CDC was calculated to be 96.65%. The GCV of the produced off-gas was calculated to be 24.64 MJ·m⁻³, which is below the effective 2H quality natural gas standards. To ensure compliance with the effective regulation, there are two key considerations:

- Recovery and reuse of H₂ component;
- Measurements need to be carried out in pressurized tank reactor;

To enhance the relatively low value of VVD, there are also two potential approaches to consider:

- Increasing carbon dioxide concentration: This can be achieved by using pure CO₂ sources or gas mixtures that contain CO₂ along with inert gas components. By increasing the CO₂ concentration in the feed gas, the availability of carbon for methanogenesis can be enhanced, leading to higher methane production rates.
- Utilizing absorbed carbon dioxide from flue gas: Carbon dioxide can be captured from flue gas emissions of industrial processes and utilized as a feedstock for biomethanation. This approach not only helps in reducing GHG emissions but also provides a sustainable source of carbon for methane production.

The integration of biomethanation P2G process with post combustion technologies offers potential advantages. However, the current challenge lies in the complexity and cost of available technologies for CCUS. The performance of these technologies and the high costs associated with regeneration hinder their widespread adoption. Simplifying the available technologies may result in drop in capital expenditures. Nevertheless, the developed or further developed CCUS concepts require novel solutions in process control. Additionally, questions regarding the installation, composition, and disposal of generated waste need to be addressed in the future to ensure a comprehensive analysis of the regulatory aspects. To gain a complete understanding of the regulation and optimize the biomethanation P2G process, further measurements need to be conducted using a pressurized tank reactor.

The commercial-scale utilization of the new process holds further potential, including:

- Evaluation of new absorbents and defining process parameters such as temperature, pressure, and concentration;
- The technology used for CCUS may be suitable for harnessing waste heat generated during the methanation process;
- Utilization of the byproduct of biogas production in integrated CCUS and P2G technologies, as well as the development of a shared heat management system;
- Further utilization of the components separated during CCUS technology for flue gas utilization.

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