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Perspectives of Biogas Plants as BECCS Facilities: A Comparative Analysis of Biomethane vs. Biohydrogen Production with Carbon Capture and Storage or Use (CCS/CCU)

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Abstract: The transition to a carbon-neutral economy requires innovative solutions that reduce greenhouse gas emissions (GHG) and promote sustainable energy production. Additionally, carbon dioxide removal technologies are urgently needed. The production of biomethane or biohydrogen with carbon dioxide capture and storage are two promising BECCS approaches to achieve these goals. In this study, we compare the advantages and disadvantages of these two approaches regarding their technical, economic, and environmental performance. Our analysis shows that while both approaches have the potential to reduce GHG emissions and increase energy security, the hydrogen-production approach has several advantages, including up to five times higher carbon dioxide removal potential. However, the hydrogen bioenergy with carbon capture and storage (HyBECCS) approach also faces some challenges, such as higher capital costs, the need for additional infrastructure, and lower energy efficiency. Our results give valuable insights into the trade-offs between these two approaches. They can inform decision-makers regarding the most suitable method for reducing GHG emissions and provide renewable energy in different settings.

Keywords: HyBECCS; hydrogen; BECCS; biogas; biohydrogen; biomethane; CDR; NET; carbon-negative hydrogen; steam methane reforming; SMR

1. Introduction

The urgent need to mitigate climate change has led to the development of innovative technologies that can reduce greenhouse gas (GHG) emissions and promote sustainable energy production. Additionally, negative emission technologies (NETs), also known as carbon dioxide removal (CDR) technologies, are crucial for limiting human-induced global warming to less than 2 °C above pre-industrial times [1,2].

Biogas, a renewable energy source produced by the anaerobic digestion of organic waste, has emerged as a promising alternative to fossil fuels [3]. During the process of biogas production, microorganisms break down organic matter in an oxygen-free environment. The result is a gas mixture that is mainly composed of methane (CH_4) and carbon dioxide (CO_2) [4]. The CH_4 contained in biogas can be purified and is then referred to as biomethane [5]. Once the biogas has been purified to methane, it can be used in various ways, e.g., as a fuel for transportation or for industrial applications. It can also be injected into and transported through the natural gas grid. In biomethane production with carbon dioxide capture and storage (CCS), the biogenic CO_2 released during the anaerobic digestion is captured and permanently stored. However, biomethane is a greenhouse gas itself

when emitted into the atmosphere, and its use in combustion engines or combined heat and power (CHP) units results in CO_2 emissions.

Alternatively, biogas can be processed by steam reforming to produce hydrogen (H_2) [6,7]. H_2 can also be used as a fuel for transportation or injected into the natural gas grid [8,9]. An advantage of H_2 over biomethane is that it can be used decentrally without emitting CO_2 or other greenhouse gases, as it is a carbon-free energy carrier. Furthermore, during the steam-methane reforming (SMR) process, in addition to the CO_2 in the biogas, biogenic CO_2 is generated as a product and can be captured and stored. This leads to a higher CO_2 removal potential compared to biomethane production with CCS. However, energy losses resulting from the conversion processes must be considered. Technologies that combine biohydrogen production with CCS are referred to as Hydrogen Bioenergy with Carbon dioxide Capture and Storage (HyBECCS) approaches [10].

Biogas reforming for hydrogen production is a promising area of research and development. Significant progress has been made in the efficiency and scalability of the technology [11–13]. Furthermore, several companies and research institutions are actively engaged in advancing biogas reforming processes, optimizing catalysts, improving reaction kinetics, and exploring their integration with CCS technologies. Some examples include the Canadian company Xebec Adsorption [14], the French Air Liquide [15], and WS Reformer in Germany [16]. Similarly, pilot projects and demonstration plants are being developed, indicating a growing interest in harnessing the potential of biogas reforming as a sustainable pathway for hydrogen production (see for example [17–19]).

For CO_2 storage, there are several options. One common method is injecting the CO_2 into underground geological formations, such as depleted oil and gas reservoirs, saline aquifers, or coal seams [2,20]. Furthermore, there are also several options for the long-term use of captured carbon dioxide (CCU) [21]. One example for CCU is the production of building materials, such as cement, concrete, and mortar, which can help reduce greenhouse gas emissions by replacing GHG-intensive materials in the building sector. As the captured and stored CO_2 is from biogenic sources, its storage or long-term use can lead to negative emissions, and the process can be considered a CDR approach or NET [22,23].

This paper compares biomethane production from biogas with CCS with biohydrogen production from biogas with CCS. The analysis considers the technical, economic, and environmental aspects of each approach. The aim is to provide insights into the trade-offs to be considered in terms of energy efficiency, their potential for negative emission generation and economic potentials. First, a general energy efficiency analysis is presented. For this, a use case is defined that involves the use of biomethane or hydrogen as a fuel in heavy-duty vehicles. Next, the negative emission potential (NEP) for each process option is calculated and compared. Finally, a discussion about the associated costs and profitability of both processes is presented.
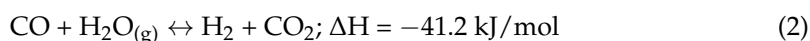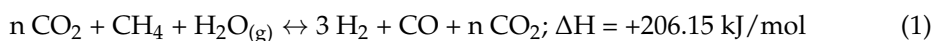
2. Basics

2.1. Biomethane Production through Biogas Purification

Biogas is produced through the anaerobic digestion of organic waste [3,24]. It consists mainly of methane (CH_4) and carbon dioxide (CO_2). Typically, the amount of CH_4 in biogas is between 55 vol% and 70 vol% [4]. Other gaseous components like hydrogen sulfide (H_2S), hydrogen (H_2), and nitrogen (N_2) can be present in small quantities. Biogas can be purified to biomethane using various technologies such as pressure swing adsorption (PSA), membrane separation, or cryogenic distillation [5,25]. The purification process removes CO_2 and impurities such as water and trace gases to increase the CH_4 content of the gas [26]. In terms of physical properties, biomethane and methane are identical, as they are the same substance. The different name only refers to their origin and production process. While "methane" refers to any type of methane, including methane derived from fossil resources, biomethane specifically designates methane obtained from biogas [27].

2.2. Biohydrogen Production through Biogas Steam Reforming

Biogas steam reforming is a process used to produce H_2 from biogas. The process involves the conversion of CH_4 into gaseous H_2 and CO_2 through a reaction with steam and the parallel occurring carbon monoxide (CO) shift. According to Rostrup-Nielsen and Christiansen, the process is described by the following two reactions [28]:

$$n\ CO_2 + CH_4 + H_2O_{(g)} \leftrightarrow 3\ H_2 + CO + n\ CO_2;\ \Delta H = +206.15\ kJ/mol \tag{1}$$

$$CO + H_2O_{(g)} \leftrightarrow H_2 + CO_2;\ \Delta H = -41.2\ kJ/mol \tag{2}$$

where n describes the molar ratio of CO_2 to CH_4 in the feed gas. This ratio is between 0.43 and 0.82, as the molar amount of CH_4 is between 55% and 70% for typical biogas plants, assuming ideal gas conditions [4]. Assuming the complete conversion of the C feedstock into CO_2, the overall stoichiometric reaction, obtained by combining Equations (1) and (2), is [29]

$$n\ CO_2 + CH_4 + 2\ H_2O_{(g)} \leftrightarrow 4\ H_2 + (n + 1)\ CO_2;\ \Delta H = +\ 164.95\ kJ/mol \tag{3}$$
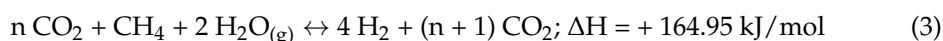
2.3. Carbon Dioxide Removal

NETs are essential to counterbalance exceedingly high GHG emissions and to limit global warming to 2 °C above pre-industrial levels [1]. NETs consist in removing CO_2 from the atmosphere and storing it for long periods, effectively reducing the overall concentration of CO_2 in the atmosphere. Negative emissions correspond to the amount of removed and stored CO_2 that exceeds the GHG emissions caused during the entire process of generating such carbon removals. This means that these GHG emissions must be deducted from the stored CO_2 to quantify the amount of negative emissions [30]. For process comparisons based on similar technologies, the maximum possible negative emissions can be estimated. For this purpose, all GHG emissions are neglected, and only the maximum captured amount of biogenic (or atmospheric) CO_2 is considered. This is a comparative value referred to as negative emission potential (NEP) [23].

CDR involves the capture and storage of atmospheric CO_2 from direct air carbon capture approaches (DACCS, direct air carbon capture and storage) and of biogenic CO_2, e.g., from the energetic use of biogenic waste (BECCS, bioenergy with carbon capture and storage). The most common method of CO_2 storage is geological storage, which involves injecting CO_2 into deep geological formations such as depleted oil and gas reservoirs, saline aquifers, and unmineable coal seams [2,31]. Another option for CO_2 storage is mineralization, which involves the reaction of CO_2 with naturally occurring minerals to form stable carbonates [32]. This process can occur naturally over long periods. However, it can be accelerated through various methods like mineral carbonation. This involves the reaction of CO_2 with magnesium and calcium silicate minerals to form stable carbonates. In addition to the permanent storage for CO_2, some carbon capture and utilization (CCU) approaches can be considered CDR, as long as the utilized carbon is durably stored in the respective product. Examples are the production of long-term materials like high-tech plastics or construction materials [33,34].

2.4. The HyBECCS Concept

HyBECCS is an umbrella term for processes in which H_2 is produced from biogenic residual and waste materials and the resulting CO_2 is captured and permanently stored. It can also be seen as a subsidiary branch of bioenergy with carbon capture and storage (BECCS) with the particularity that H_2 is produced as an energy source. The technical approach can be split into four basic process steps: 1. substrate biomass pretreatment, 2. the production of biohydrogen and biogenic CO_2, 3. the separation of the product gases CO_2 and H_2, and 4. their processing for the use of H_2 and long-term storage or the use of biogenic CO_2, including the permanent CO_2 storage or use itself [10]. For each of the four

steps, there are different technology options. A main technological advantage of BECCS and HyBECCS approaches is the energy-efficient capture of biogenic CO_2 as it generally occurs as a point source. It provides a double effect on climate mitigation by providing GHG-free energy and the storage of biogenic CO_2 with the potential to provide negative emissions. The basic requirements are, however, the deduction of all GHG emissions occurring over the entire process chain to determine the amount of negative emissions, economic viability, and a reliable long-term CO_2 storage [30]. Considering biogas plants, there are several possibilities to retrofit existing plants to HyBECCS facilities [35]. The technical approach considered in this work combines biogas-steam reforming with CCS, as described in the following section.

3. Process Descriptions and Main Assumptions

This section defines and describes the two process options to be compared in this work: biomethane production with CCS (process option 1) and biohydrogen production through biogas-steam reforming with CCS (process option 2). The various stages of each process option with a focus on the differences are described and the main assumptions for their comparison are outlined.

An overview of the two process options is provided in Figure 1. The first stage of both process options involves the preparation of the feedstock, followed by biogas production through the anaerobic digestion of organic waste in large vessels that provide an oxygen-free environment for microorganisms to break down the organic matter [3]. Different types of organic waste can be used, including agricultural waste, food waste, and sewage [36,37].
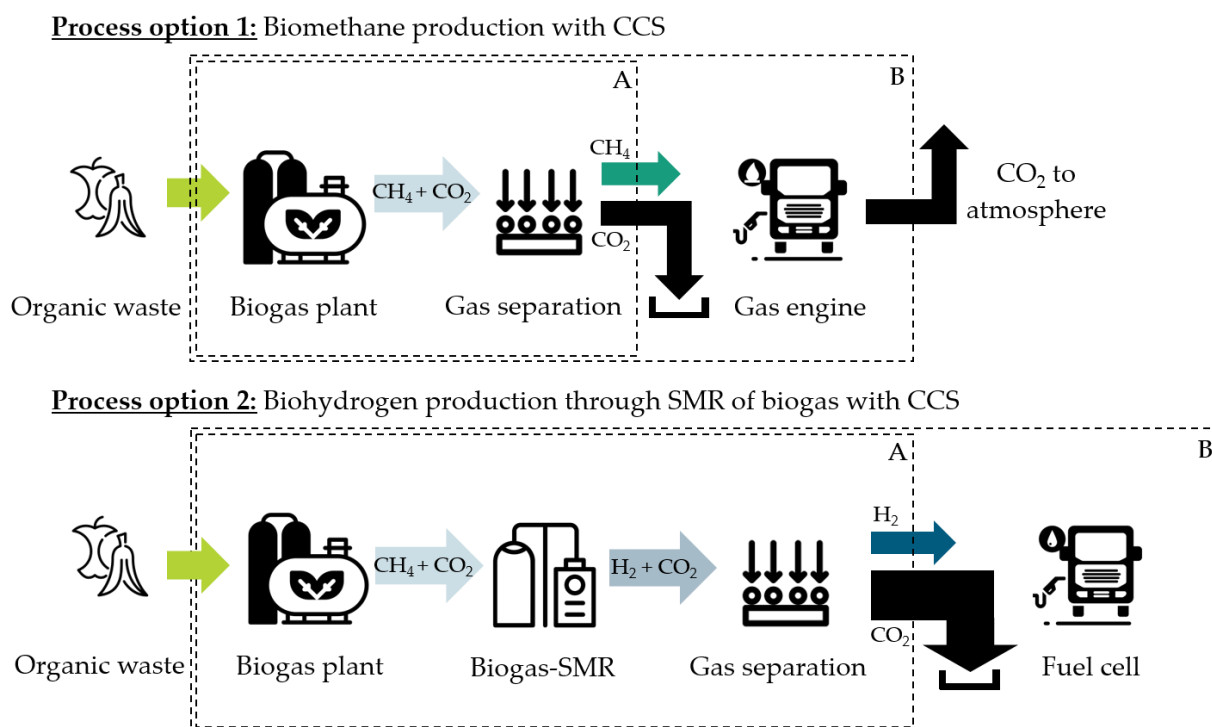
Process option 1: Biomethane production with CCS

Process option 2: Biohydrogen production through SMR of biogas with CCS

Figure 1. Process options and system boundaries (A and B) for biogas usage (own representation).

In process option 1, the second stage is separating the CH_4 from the other gas components in the biogas. This is typically performed by PSA that uses adsorbent materials to selectively adsorb CO_2 and impurities, leaving behind purified biomethane, as described in Section 2.1 [25,27,38]. Alternatively, membrane separation or water scrubbing can be used.

For process option 2, the biogas is fed into a steam reforming process, as described in Section 2.2. The resultant gas mixture is subjected to further treatment, including a water-gas shift to increase the H_2 and CO_2 concentration, and gas separation through PSA

to gain the product fractions CO_2 and H_2. Relevant reaction equations for this process option are derived below.

The next step for both process options is CO_2 capture and storage. In process option 1, the biogenic CO_2 from the biogas supply is captured after the separation of the biomethane. In process option 2, biogenic CO_2 is present in the biogas fuel and feed and enriched during biogas burning, reforming and CO-shift steps. After the H_2 separation, the tail gas is redirected to the burner to be oxidized, resulting in further CO_2. The aggregated CO_2 can be captured from a single exhaust gas stream. The essential difference between the two process options is that in process option 1, only the CO_2 present in the biogas can be captured. Part of the carbon content of the original biomass is bound in the CH_4 and is released during the decentralized use of the methane. In contrast, in process option 2, the entire carbon content of the biogas is released during the SMR step and can be captured from the "Exhaust Gas" stream (cf. Figure 2) as CO_2.
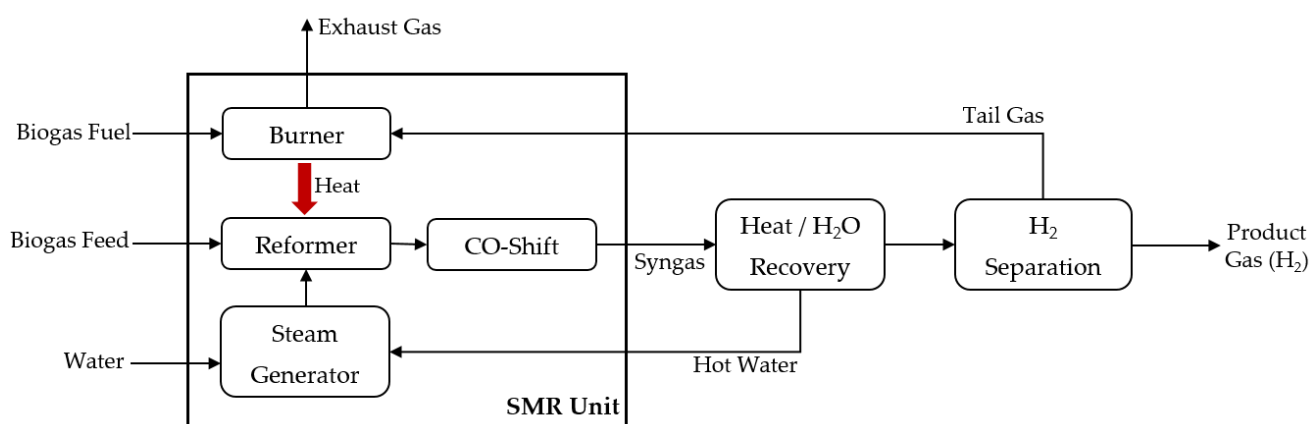

Figure 2. Flow diagram of the steam methane reforming process step within process option 2 (own representation).
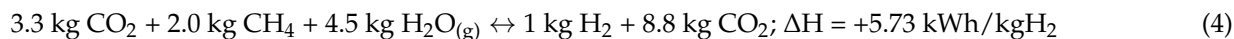
After its separation, the CO_2 is compressed for transportation either via pipelines or in containers by truck. Storage sites are often located offshore, which may require additional transport by ship. Which option is more economically and environmentally viable must be decided for each case specifically. Transport over short distances can be carried out by truck and ship, while transport via pipeline involves higher expenditures, but becomes viable in the long term if CO_2 is transported over long distances and when the amounts of CO_2 transported are higher [39,40]. Finally, the CO_2 is stored to prevent its emission into the atmosphere, e.g., in geological formations, such as depleted oil and gas reservoirs or saline aquifers [2].

It is assumed that the use of both chemical energy carriers H_2 and CH_4 provided in process options 1 and 2 is decentralized and that no carbon capture occurs during their use. For example, vehicles can be fueled, or heat can be supplied. Two cases are defined for exemplification: Case A (see system boundary A in Figure 1) compares the biomethane from process option 1 to the biohydrogen produced via process option 2 in terms of their chemical energy content as secondary energy carriers, expressed by the lower heating value (LHV). As depicted in Figure 1, the system boundaries for case A range from the biomass supply to the biomethane and H_2 production.

Case B (see system boundary B in Figure 1) compares the useful energy generation potentials of both process options for heavy-duty transport applications. In process option 1, the useful energy from biomethane is generated by bio-liquefied natural gas (LNG) combustion engines. In process option 2, the energy from H_2 is generated with fuel cell power trains. Hence, the system boundary B is set from the biomass supply to the use of biomethane and H_2, respectively, for heavy-duty transportation (system boundary B in Figure 1). Burning liquefied biomethane in gas engines can reach efficiencies up to 44% [41]. State-of-the-art fuel cell systems for electric truck powertrains peak at efficiencies
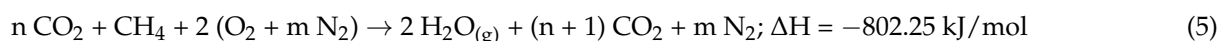
of 63% [42–44]. In the following, with regard to further technological development and optimization, high efficiencies of 44% (for bio LNG engines) and 63% (for H_2 fuel cell engines) are assumed for both process options.

Relevant reaction equations for Process option 2 are derived in the following: An average biogas composition of 37.5 vol% CO_2 and 62.5 vol% CH_4 is assumed [4]. By converting the units and referring to the product of 1 kg H_2, reaction Equation (3) can be written as follows:

$$3.3 \text{ kg } CO_2 + 2.0 \text{ kg } CH_4 + 4.5 \text{ kg } H_2O_{(g)} \leftrightarrow 1 \text{ kg } H_2 + 8.8 \text{ kg } CO_2; \Delta H = +5.73 \text{ kWh/kgH}_2 \qquad (4)$$
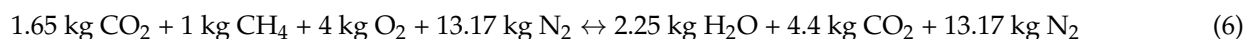
This reaction is endothermic and, thus, energy must be provided to drive the reaction towards H_2 and CO_2 production Typically, this energy is supplied in the form of heat, generated by the oxidation of a part of the biogas and of the tail gas after H_2 separation. Figure 2 shows the schematic structure of the SMR process with the respective material and energy flows.

As depicted in Figure 2, the ΔH in reaction (3) representing the endothermic heat of the reaction has to be provided by the burner being part of the SMR unit. Additionally, there is heat demand for steam generation and preheating the feedstock to typical operation temperatures in the reformer of around 800 °C, as well as for compensating wall heat losses and latent heat of the products. The total heat energy supplied amounts to approx. 25 kWh per kg of H_2 produced. This value is based on empirical data from existing biogas SMR units and is taken as an assumption in this paper. The combustion of biogas with air is described by the following equation:

$$n \text{ } CO_2 + CH_4 + 2 \text{ } (O_2 + m \text{ } N_2) \rightarrow 2 \text{ } H_2O_{(g)} + (n + 1) \text{ } CO_2 + m \text{ } N_2; \Delta H = -802.25 \text{ kJ/mol} \qquad (5)$$

where n describes the molar ratio of CO_2 to CH_4 in the feed gas and m describes the ratio of N_2 to O_2 in air. Usually air contains around 79 vol% N_2 and 21 vol% O_2 [45]. By converting the units and referring to 1 kg CH_4, Equation (5) can be rewritten as follows:

$$1.65 \text{ kg } CO_2 + 1 \text{ kg } CH_4 + 4 \text{ kg } O_2 + 13.17 \text{ kg } N_2 \leftrightarrow 2.25 \text{ kg } H_2O + 4.4 \text{ kg } CO_2 + 13.17 \text{ kg } N_2 \qquad (6)$$

It can be seen that 62.5% of the CO_2 in the exhaust gas originates from the combustion of methane, while 37.5% comes from the biogas.

Summing up, the two process options consist of several identical process steps but also show some differences: The preparation of the feedstock, the biogas production in the biogas plant and the product gas treatment are identical for both processes. To compare both processes from an ecological point of view, these identical process steps can be excluded from the comparison according to the so-called "black-box" approach [46,47]. Also, for the economic comparison, identical plant components can be neglected.

The main differences between the two process options are: (i) The additional biogas SMR step in process option 2. (ii) The higher amount of captured CO_2 leading to more CO_2 to be compressed, transported, and stored in process option 2. Both (i) and (ii) increase the investment and operational costs of process option 2. (iii) The different products with their respective technology options for heavy-duty transportation: biomethane and biogenic CO_2 in process option 1 versus biohydrogen and more biogenic CO_2 in process option 2. Further differences are the technological and cost differences between H_2 and biomethane in compression and transportation.

4. Technology Comparison

4.1. Considerations on Product Application

In this section, the two products, H_2 and CH_4, are compared regarding their product properties. This comparison is independent of the two use cases described in Section 3 and intends to give a broader view on possible applications.

CH_4 has a higher volumetric energy density than H_2, which means it contains more energy per unit volume [48]. This makes CH_4 generally more suitable for applications where space is limited. However, H_2 has a higher energy content per unit mass, which

makes it more efficient for applications where weight is a concern [49]. CH_4 and H_2 are both flammable and explosive above certain concentrations [50]. However, H_2 has a wider flammability range and can ignite at lower concentrations. With proper handling and storage, both gases can be used safely [51].

Both biomethane and H_2 are considered to play key roles in future energy provision, e.g., as substitutes for natural gas [52,53]. They can be used for transportation, heating, power generation, and in industrial processes, as described briefly in the following.

CH_4 is commonly used as a fuel for vehicles, especially in the form of compressed natural gas (CNG) or LNG [54,55]. CNG is used in light-duty vehicles such as cars, while LNG is used in heavy-duty vehicles like trucks and buses [56,57]. H_2 can be used with fuel-cell vehicles (FCVs) as well as modified internal combustion engine vehicles [58]. Furthermore, CH_4 as well as H_2 can be used for heating and power generation in residential, commercial and industrial applications. H_2, however, generally requires a specific environment for combustion due to its lower volumetric energy density, faster laminar flame speed, and higher combustion temperatures (1600 °C) compared to CH_4 (1300 °C) [59]. Subsequently, conventional gas engines must first be either retrofitted or replaced with gas turbines that are H_2-ready and thus adequate for the use of 100% H_2.

4.2. Energy Efficiency

Comparing process options 1 and 2, as described in Section 3, the chemical energy contents of the produced gases are a critical factor for energy efficiency comparison. Therefore, the available thermal energy produced by a combustion of the gases, expressed by the mass-specific lower heating value (LHV), was considered. As described in Section 2.2, the reactions of biomethane to CO_2 and H_2 proceed under energy input. The thermal energy for the endothermic reaction is obtained via the oxidation of biogas. Since CO_2 is completely oxidized, the thermal energy is obtained via full oxidation of the CH_4 in the biogas.

For the following calculation, it is further assumed that biogas consists of 62.5 vol% CH_4 and 37.5 vol% CO_2. This is an average value based on existing biogas processes, as described in Section 2.2. Equation (7) is derived from Equation (4) under consideration of the 25 kWh thermal energy input required for the overall SMR process, as described in Section 3.

$$2.0 \text{ kg CH}_4 + 4.5 \text{ kg H}_2\text{O}_{(g)} + 25 \text{ kWh}_{th} \rightarrow 1.0 \text{ kg H}_2 + 5.5 \text{ kg CO}_2 \tag{7}$$
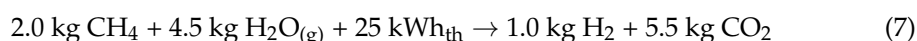
The chemical energy content of CH_4 and H_2 was calculated with Equation (8) using the LHV for the quantities given. The LHV of CH_4 is 13.9 kWh/kg and of H_2 33.33 kWh/kg [60].

$$E_i = \text{LHV} \times m_i \tag{8}$$

E_i is the total chemical energy content for the considered mass of the species i [in kWh] and m_i the mass of the species i [in kg]. In this case, 2.0 kg of CH_4 has an energy content of 27.8 kWh. Thus, 52.8 kWh of energy input, divided into 2.0 kg CH_4 (LHV) and thermal energy of 25 kWh, leads to 33.33 kWh of chemical energy produced in the form of 1 kg H_2 (LHV), as shown in Table 1. Therefore, the efficiency of the considered steam-reforming process is 63% in terms of the chemically stored energy content of the product H_2 in relation to the overall energy input.

Table 1. Energy balance for the production of 1.0 kg H_2 by biogas steam reforming (process option 2, system boundary A).

	Energy Input	Energy Output
Chemical energy content CH_4	27.8 kWh	0.0 kWh
Chemical energy content H_2	0.0 kWh	33.33 kWh
Thermal energy	25.0 kWh	0.0 kWh
Total	52.8 kWh	33.33 kWh

To compare the energy efficiency of CH_4 and H_2 in the heavy-duty transportation sector (case B), the efficiencies of the corresponding technologies must be included. As listed in Section 3, gas engines for liquefied biomethane can reach efficiencies up to 44%, while fuel cell systems with batteries for H_2 peak up to 63% [42–44]. The efficiencies can be included using the following equation.

$$E_{in} = E_{out}/\eta \tag{9}$$

where E_{in} is the energy in kWh, E_{out} is the usable energy in kWh and η is the efficiency of the corresponding technology.

For biomethane, providing 1 kWh of usable energy for transport requires an energy input of 2.27 kWh at an efficiency of 44%. For H_2, the calculation must also take into account the energy efficiency of its production via steam reforming with a value of 63.1%, as shown above and in Table 2. Thus, for a 63% efficiency of the fuel cell system with a battery, the total efficiency of process option 2 for case B is 39.8%. This means that to supply 1 kWh of usable energy in the form of H_2, an energy input of 2.52 kWh is needed. Hence, using H_2 produced in process option 2 means a loss of approx. 4.2% of the usable energy in the form of propulsion energy for heavy-duty transport compared to process option 1.

Table 2. Required energy input for the production of 1.0 kWh usable energy (process options 1 and 2, system boundary B).

	Process Option 1	Process Option 2
Usable Energy	1.0 kWh	1.0 kWh
Efficiency (heavy-duty transport)	44.0%	63.0%
Relative Efficiency (production)	100%	63.1%
Total Efficiency	44.0%	39.8%
Energy Input	2.27 kWh	2.52 kWh

However, this is within the range of several uncertainties due to the assumptions made, e.g., assumed powertrain efficiencies and different driving behaviors. Furthermore, the compression of the two gases, CH_4 and H_2, are not considered. To convert CH_4 gas into LNG, an energy loss of 10–20% can be assumed (1.39–2.78 kWh/kg CH_4) [61]. For H_2 compression to 400 bar, an energy loss of about 10.8% (3.61 kWh/kg H_2) can be assumed [62]. Since the energy consumptions for both applications are associated with uncertainties and are in a similar order of magnitude, they can be neglected in the calculations.

In conclusion, however, it can be said that for Use Case B, the efficiency losses that occur in process option 2 due to the reforming process step (Use Case A) can be counterbalanced by better efficiency in the FCEV power train, leading to the useful energy for heavy-duty transport being in a similar order of magnitude for both process options.

4.3. Negative Emission Potential

The NEP represents the amount of negative emissions that are possible if all GHG emissions caused along the entire value chain up to the use or storage of the product gases are neglected, as described in Section 2.3. It can be used to compare the two process options described in Section 3 following the "black box" approach, where identical process steps are excluded from the comparison [46,47]. To determine the NEP, the theoretical maximum amount of storable biogenic CO_2 is calculated for the process options. Therefore, the amount of CO_2 generated via biogas production in the biogas plant is considered first. It is assumed, as described in Section 4.2, that biogas consists of CH_4 (62.5 vol%) and CO_2 (37.5 vol%). Thus, 1 Nm^3 biogas consists of 0.375 m^3 CO_2 and 0.625 m^3 CH_4. Furthermore, the biogas was considered an ideal gas. According to the ideal gas law [63], this leads to the following equation being applicable:

$$pV_k = n_k RT, \tag{10}$$

where p is the pressure in Pascal, V_k is the volume of the species k in m^3, n_k is the amount of the species k in mol, R is the general gas constant (in J/mol·K), and T is the temperature in K. From the ideal gas law, a molar ratio of 0.6 mol CO_2/mol CH_4 is obtained for the assumed average composition of biogas. This corresponds to a mass ratio of 1.65 kg CO_2/kg CH_4 and means that 1.65 kg of CO_2 is produced via the biogas plant per kilogram of CH_4 produced in process option 1. In process option 2, an additional 2.75 kg of CO_2 is produced from heat supply through biogas combustion and reforming, when the same amount of biogas containing 1 kg of CH_4 is supplied as input stream. Adding this amount to the biogenic CO_2 in the biogas leads to a total of 4.4 kg CO_2 to be captured in process option 2.

In summary, the biomethane production (process option 1) results in an NEP of 1.65 kg CO_2 per kg CH_4 produced, while the HyBECCS approach (process option 2) shows a NEP of 4.4 kg CO_2 for the same biomass input. This corresponds to about 2.7 times the amount of storable biogenic CO_2 of process option 2 compared to process option 1. Furthermore, depending on the biogas quality (CH_4 content), this ratio deviates, as shown in Figure 3. The NEP of process option 2 increases to up to 3.3 times the amount of process option 1 for a CH_4 content in the biogas of 70 vol%. For a CH_4 content in biogas of 55 vol%, the NEP for process option 1 is still 2.2 times higher than that of process option 1.
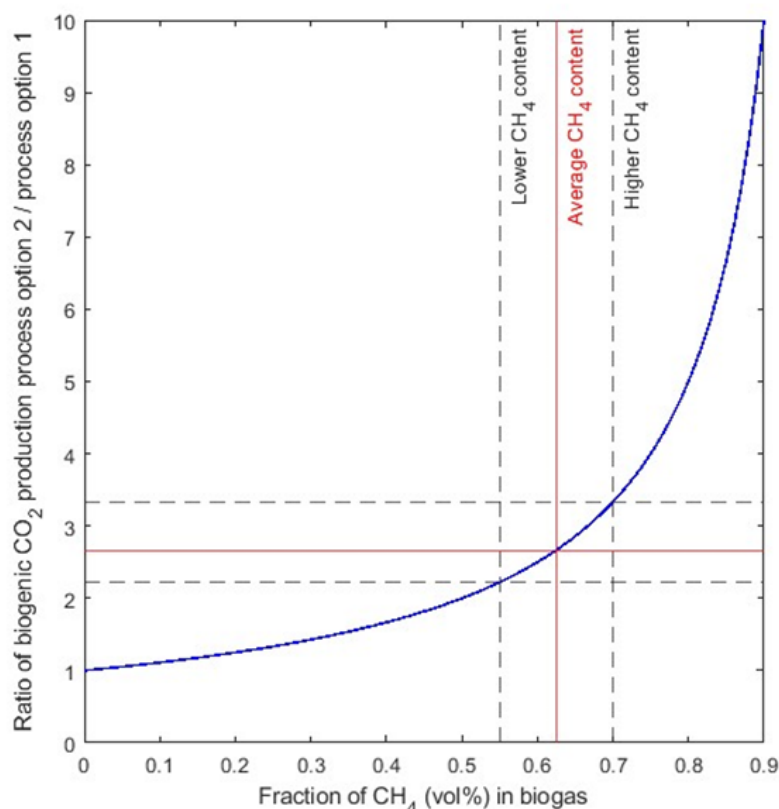

Figure 3. Ratio of biogenic CO_2 production of process option 2 vs. process option 1 for different CH_4 volume fractions in biogas (own representation).

4.4. Economic Comparison

The costs required and revenues to be expected for the two process options can vary significantly depending on the specific technology and scale of the project. However, some general considerations can be made.

Costs that arise in both process options can be neglected in the comparison, e.g., the main technologies used for upgrading biogas to biomethane are PSA and membrane separation [64]. This is also required for the purification of the biohydrogen and can therefore be neglected for the comparison [65]. Additionally, there may be costs associated with obtaining permits, certifications, and approvals for the biomethane as well as the

biohydrogen production, depending on the regulations in the specific country or region. These costs can also vary depending on the project location and regulatory environment and are neglected in this work.

The costs for biogas steam reforming to produce H_2 include the capital costs of the reforming unit, which is the main equipment required for the process. Therefore, the capital costs for process option 1 are generally lower than for process option 2. This is primarily because additional plant components for the reforming process step are needed in process option 2. The costs for the SMR upgrade have to be analyzed specifically for each case. However, the additional costs for the SMR upgrade have to be compensated by a higher income from the sale of the products in order to be economically feasible. The income side is described in the following.

Furthermore, the different sources of income with their respective market prices, today and in the future, can be compared. In process option 1, the marketable products are CH_4 and negative emissions, whereas, in process option 2, the marketable products are H_2 and negative emissions. The total income for each process option depends on the market prices of the respective products today and in the future during the lifetime of the plant as well as the generated amount of each product. A total of 3.6 kg of CH_4 is necessary for the production of 1 kg of H_2, as described in Section 3. Process option 2 therefore requires 3.6 times the amount of biogas input per kg of produced energy carrier. Considering negative emissions, however, process option 2 has the potential to produce about 2.7 times the amount of storable biogenic CO_2 to be captured and stored, as shown in Section 4.3.

The current market prices of CH_4 and H_2 vary depending on the region and quality. However, some general price comparisons can be made based on available data. In the United States, the retail price of CNG for transportation ranges from USD 2–4 per gasoline gallon equivalent (GGE) (EUR 0.72–1.45 per kg natural gas [66]), whereas the retail price of LNG for transportation ranges from USD 2–5 per diesel gallon equivalent (DGE) (EUR 0.68–1.69 per kg LNG [66]), both depending on the location and the supplier. The retail price of H_2 for transportation ranges from USD 9.5–13.2 per kg (EUR 8.8–12.3 per kg), depending on the location and the supplier [67]. In Europe, the fuel prices for transportation vary from country to country. In the first quarter of 2022, the average fuel price range for CNG in Europe was EUR 5.23 to EUR 12.51 per 100 km travelled (average EUR 7.24 per 100 km), based on information submitted by nine member states [68]. Assuming an average consumption of 3.64 kg of CNG per 100 km [69], this corresponds to a price range of EUR 1.44 to EUR 3.43 per kg of CNG (Average: EUR 1.99 per kg). For LNG, the average retail price range for the same period was from EUR 0.45 to EUR 2.81 per kg of LNG, with an average price for Europe (EU-27) of EUR 1.86 per kg LNG [70]. With an average consumption of 2.14 kg LNG per 100 km for heavy trucks [56], this corresponds to a price range of EUR 0.96 to EUR 6.01 per 100 km (average: EUR 3.98 per 100 km). Average H_2 prices in the first quarter of 2022 ranged from EUR 9.00 to EUR 15.06 per 100 km (average: EUR 11.22 per 100 km), based on information submitted by six member states. Assuming an average consumption of 0.8 kg H_2 per 100 km [71], an average price range of EUR 11.25 to EUR 18.83 per kg of H_2 (average: EUR 14,01 per kg H_2) is obtained. An overview of the pump price ranges for different fuels in the European Union is shown in Table 3.

Table 3. Overview of price ranges for different fuels in the European Union (average for Q1 of 2022) [68,70].

Fuel	Price (EUR/kg)	Price (EUR/100 km)	Reference
CNG	1.44–3.43 Average: 1.99	5.23–12.51 Average: 7.24	[68]
LNG	0.45–2.81 Average (EU-27): 1.86	0.96–6.01 Average (EU-27): 3.98	[70]
H_2	11.25–18.83 Average: 14.01	9.00–15.06 Average: 11.22	[68,71]

Summarizing, the current market prices of CH_4 are clearly lower than those of H_2. This is due to H_2 being a downstream product predominantly produced from CH_4 obtained from natural gas [72]. However, the long-term price forecasts for H_2 indicate potential for lower prices due to cost reductions through technological alternatives like electrolysis as well as economies of scale [73].

Both process options generate negative emissions, when the biogenic CO_2 is not released into the atmosphere but captured, transported to a storage site, and permanently stored. However, generating negative emissions from biogenic CO_2 creates further costs and will, therefore, only happen when they are overcompensated by an income. One option is the sale of so-called carbon removal units (CRUs) issued for verified negative emissions (in tCO_2e) and can be sold to buyers who wish to claim them. Already today, several private entities offer CRUs [74]. They stem from different carbon-removal approaches in different countries with different underlying quality standards and show broad price ranges. Examples are EUR 20/CRU for bio-based construction materials in France to EUR 535/CRU for biochar in Sweden [75]. In November 2022, the European Commission proposed an EU carbon-removal certification framework (CRCF) to establish common rules for the monitoring, verification, and reporting of negative emissions for the voluntary carbon removal market [76]. This EU initiative is still ongoing. However, this standardization process within the geographical boundaries of one of the biggest carbon trading schemes worldwide shows the awareness of the political bodies for the urgent need to create a secure and credible playing field for the commercialization of CRUs. Summing up, negative emissions are already today a marketable product, and, in the future, ongoing standardization processes will increase the visibility, reliability, and, thus, the market volume for negative emissions in Europe and worldwide.

As outlined in Section 4.3, process option 1 results in an NEP of 1.65 kg CO_2 per kg of CH_4 produced, while process option 2 shows an NEP of 4.4 kg CO_2 for the same amount of biomass input. The NEP represents the maximum amount of negative emissions to be produced and qualified for CRUs in each option. Process option 1 generates 1.0 kg of bio-LNG, while process option 2 generates 0.5 kg H_2 for the same amount of biomass input, as outlined in Section 3. The following price ranges, also described above, give the following price averages for the products: For H_2, the price range of 0.93–3.72 EUR/kg H_2 results in an average price of 2.325 EUR/kg H_2. For LNG, the price range of 0.68–1.69 EUR/kg LNG results in an average price of 1.185 EUR/kg LNG. For CRU, the price range of 20–535 EUR/CRU results in an average price of 277.5 EUR/CRU. For process option 1, this means that the products (LNG and CO_2) can generate an income of 459.06 EUR/kg of CH_4 produced. For process option 2, the products (H_2 and CO_2) can generate an income of 1222.16 EUR for the same amount of biomass input. In this simplified exemplary calculation, process option 2 would generate more income. However, cost consideration is necessary for final economic comparison and CRU pricing is still not fixed or legislated. The calculation shows, however, that depending on the market value of the different products, yields determine the superiority in terms of income of the process options and gives first indications and orientation values.

5. Limitations

This paper focuses on comparing the biomethane vs. biohydrogen with CCS options in terms of their energy efficiency, economic aspects, and negative emission potential. Other environmental impacts are not considered. However, both approaches' impacts on the environment must be analyzed for a holistic assessment. Impacts on social sustainability are neglected in this paper.

The energy efficiency for the steam reforming of biogas was calculated under the assumption that an additional 25 kWh of thermal energy is required for the process. This includes, on the one hand, the reformation energy but also, for example, the energy required to heat the gases or to evaporate the liquid water. Depending on the ambient parameters or energetic optimizations, the value for the required thermal energy can be higher or lower,

resulting in a change in energy efficiency. This assumption is based on experience values and should be validated. This also applies to all other assumptions, such as the efficiency of the LNG or fuel-cell propulsion and the compression/cooling of the gases in Use Case B or the average biogas composition.

Considering the economic comparison and the wide price ranges, as well as the different cost structures of both process options, requires a deeper economic analysis, taking into consideration more influencing factors such as investment and operational costs, location, and the time of operational initiation of the plant.

6. Summary and Conclusions

Climate change constitutes one of the most pressing challenges of our times. Innovative technologies are needed that can reduce GHG emissions in all sectors. Action is especially necessary in the energy sector, which is currently responsible for the largest amount of emissions. A transition to cleaner energy sources like renewable energies and carbon-free energy carriers like H_2 is thus urgently needed. Additionally, to limit human-induced global warming to less than 2 °C compared to pre-industrial levels, NETs or CDR technologies are indispensable.

In this paper, biomethane production from biogas with CCS (process option 1) was compared to biohydrogen production via the SMR of biogas with CCS (process option 2), i.e., a HyBECCS approach. The analysis considers technical, economic, and environmental aspects of the two options. The aim was to provide insights into the trade-offs to be considered.

Considering energy efficiency, the production of biohydrogen in process option 2 results in an energy loss of approximately 37% in terms of the absolute LHV of the products per kg of biomass or biogas input. Looking at usable driving energy for heavy-duty transport applications, using H_2 produced in process option 2 still results in a comparable loss of approx. 4.2% in terms of usable driving energy for the heavy-duty transport sector. However, this is within the range of several uncertainties due to the assumptions made, e.g., assumed powertrain efficiencies, LNG liquefaction (10–20% energy loss), and different driving behaviors. In conclusion, the useful energy generation for heavy-duty transport can be expected to be in a similar order of magnitude for both process options.

To determine the negative emission potential, the theoretical maximum amount of storable biogenic CO_2 (NEP) was calculated for both process options. In comparison, process option 1 results in an NEP of 1.65 kg CO_2 per kg of CH_4 produced, while the HyBECCS approach (process option 2) shows an NEP of 4.4 kg CO_2 for the same amount of biomass input. This corresponds to about 2.7 times the amount of storable biogenic CO_2. Depending on the quality of the biogas process step (CH_4 content in the biogas), even more biogenic CO_2 can be captured and stored with the HyBECCS approach compared to process option 1. The NEP of process option 2 increases to up to 5 times the amount of process option 1 for a CH_4 content in biogas of 70 vol%. For a CH_4 content of biogas of 55 vol%, the NEP for process option 2 is still 2.2 times higher than that of process option 1.

The costs required for the two process options can vary significantly depending on the specific technology and scale of the reforming plant considered. Biogas SMR requires additional equipment leading to higher investment costs. Further to this, the current market prices for CH_4 are generally lower than for H_2. However, the long-term price forecasts for H_2 indicate a potential for lower prices due to cost reductions through technological advances and economies of scale. Negative emissions are a marketable product already. In the future, ongoing standardization processes will increase their visibility, reliability, and, thus, market potentials in Europe and worldwide. This market potential correlates with the NEP and is thus about 2.7 times higher for process option 2.

In conclusion, producing biohydrogen via the SMR of biogas offers the possibility to capture between 2.2 and 5 times more of the carbon bound in biomass compared to biomethane production. Captured biogenic CO_2 has the potential to create negative emissions that can be marketed as a byproduct besides the resulting energy carrier. However, the

production of biohydrogen from biomethane results in an energy loss of approximately 37% in terms of the LHV of the product gases, but this loss can likely be partially compensated by efficient driving technologies such as FCEVs.

7. Outlook

Negative emissions will gain relevance in the coming decades, further highlighting the benefits of HyBECCS approaches such as process option 2 in this paper. Overall, they have the potential to make a significant contribution to lowering GHG emissions and establishing CO_2 sinks, thereby making the energy sector more sustainable. A deep economic analysis, considering several influencing factors such as investment and operational costs, product price forecasts, scale, location, and the time of a plant's operational initiation, must be carried out for a thorough comparison of the presented process options. Furthermore, the ecological impact of both process options has to be analyzed in a holistic assessment, including the GHG emissions caused throughout the entire lifecycle of each product couple. Therefore, developing an approach to determine the positive and negative impacts on the climate to identify the approach with the highest climate-change mitigation potential is required. This includes the development of an approach to evaluate different CCS and CCU options concerning their respective GHG mitigation potential. To allow for comparisons with other HyBECCS and NETs, the central Key Performance Indicators (KPIs) of leveled costs of carbon-negative H_2 (LCCNH) and leveled costs of negative emissions (LCNE) must be calculated for both process options [30]. Comparing the KPIs is essential for cost efficiency in NET and HyBECCS development and deployment.

Market integration of HyBECCS plants depends on establishing infrastructure for transporting both H_2 and CO_2. The different options and their future perspectives must be analyzed considering their influence on the feasibility and economics of HyBECCS approaches. Furthermore, the feasibility of specific HyBECCS plants depends on the market for both products. Thus, analyzing the present and future market potential of H_2 and CO_2 in selected geographical areas is needed.

References

1. IPCC. *Global Warming of 1.5 °C: IPCC Special Report on Impacts of Global Warming of 1.5 °C above Pre-industrial Levels in Context of Strengthening Response to Climate Change, Sustainable Development, and Efforts to Eradicate Poverty*; Cambridge University Press: Cambridge, UK, 2022; pp. 1–24. [CrossRef]
2. IPCC. *IPCC Special Report on Carbon Dioxide Capture and Storage*; Cambridge University Press: Cambridge, UK; New York, NY, USA, 2005; ISBN 13 9780521866439.
3. Weiland, P. Biogas production: Current state and perspectives. *Appl. Microbiol. Biotechnol.* **2010**, *85*, 849–860. [CrossRef]

4. Awe, O.W.; Zhao, Y.; Nzihou, A.; Minh, D.P.; Lyczko, N. A Review of Biogas Utilisation, Purification and Upgrading Technologies. *Waste Biomass Valor.* **2017**, *8*, 267–283. [CrossRef]

5. Ryckebosch, E.; Drouillon, M.; Vervaeren, H. Techniques for transformation of biogas to biomethane. *Biomass Bioenergy* **2011**, *35*, 1633–1645. [CrossRef]

6. Hajjaji, N.; Martinez, S.; Trably, E.; Steyer, J.-P.; Helias, A. Life cycle assessment of hydrogen production from biogas reforming. *Int. J. Hydrogen Energy* **2016**, *41*, 6064–6075. [CrossRef]

7. Kumar, R.; Kumar, A.; Pal, A. Overview of hydrogen production from biogas reforming: Technological advancement. *Int. J. Hydrogen Energy* **2022**, *47*, 34831–34855. [CrossRef]

8. Shabani, B.; Andrews, J. Hydrogen and Fuel Cells. In *Energy Sustainability through Green Energy*; Springer: New Delhi, India, 2015; pp. 453–491. [CrossRef]

9. Sharma, A.; Kumar Kar, S. *Energy Sustainability through Green Energy*, 1st ed.; Springer: New Delhi, India, 2015; ISBN 9788132223375.

10. Full, J.; Merseburg, S.; Miehe, R.; Sauer, A. A New Perspective for Climate Change Mitigation—Introducing Carbon-Negative Hydrogen Production from Biomass with Carbon Capture and Storage (HyBECCS). *Sustainability* **2021**, *13*, 4026. [CrossRef]

11. Nahar, G.; Mote, D.; Dupont, V. Hydrogen production from reforming of biogas: Review of technological advances and an Indian perspective. *Renew. Sustain. Energy Rev.* **2017**, *76*, 1032–1052. [CrossRef]

12. Gao, Y.; Jiang, J.; Meng, Y.; Yan, F.; Aihemaiti, A. A review of recent developments in hydrogen production via biogas dry reforming. *Energy Convers. Manag.* **2018**, *171*, 133–155. [CrossRef]

13. Rosha, P.; Rosha, A.K.; Ibrahim, H.; Kumar, S. Recent advances in biogas upgrading to value added products: A review. *Int. J. Hydrogen Energy* **2021**, *46*, 21318–21337. [CrossRef]

14. Ivys Adsorption Inc. Xebec. Available online: https://xebecinc.com/ (accessed on 19 June 2023).

15. Air Liquide. A World Leader in Gases, Technologies and Services for Industry and Health. Available online: https://www.airliquide.com/ (accessed on 19 June 2023).

16. WS Reformer GmbH. X-to-Hydrogen. Available online: https://wsreformer.de (accessed on 19 June 2023).

17. European Commission. Advanced Direct Biogas Fuel Processor for Robust and Cost-Effective Decentralised Hydrogen Production: Using Renewable Biogas to Produce Green Hydrogen. 2021. Available online: https://cordis.europa.eu/article/id/435357-using-renewable-biogas-to-produce-green-hydrogen (accessed on 19 June 2023).

18. European Commission. BIOgas Membrane Reformer for Decentralized Hydrogen Production. Available online: https://doi.org/10.3030/671459 (accessed on 19 June 2023).

19. Deutsches Biomasseforschungszentrum. BioH2Ref—Dezentrale Wasserstoffaufbereitung von Biogas durch Dampfreformierung. 2023. Available online: https://www.energetische-biomassenutzung.de/projekte-partner/details/project/show/Project/BioH2Ref-706 (accessed on 19 June 2023).

20. Mirza, N.; Kearns, D. State of the Art: CCS Technologies 2022. 2022. Available online: https://www.globalccsinstitute.com/wp-content/uploads/2022/05/State-of-the-Art-CCS-Technologies-2022.pdf (accessed on 21 April 2023).

21. Aresta, M. *Carbon Dioxide as Chemical Feedstock*; Wiley-VCH: Weinheim, Germany, 2010; ISBN 9783527324750.

22. Kemper, J. Biomass and carbon dioxide capture and storage: A review. *Int. J. Greenh. Gas Control* **2015**, *40*, 401–430. [CrossRef]

23. Full, J.; Trauner, M.; Miehe, R.; Sauer, A. Carbon-Negative Hydrogen Production (HyBECCS) from Organic Waste Materials in Germany: How to Estimate Bioenergy and Greenhouse Gas Mitigation Potential. *Energies* **2021**, *14*, 7741. [CrossRef]

24. Strevett, K.A.; Vieth, R.F.; Grasso, D. Chemo-autotrophic biogas purification for methane enrichment: Mechanism and kinetics. *Chem. Eng. J.* **1995**, *58*, 71–79. [CrossRef]

25. Abd, A.A.; Othman, M.R.; Majdi, H.S.; Helwani, Z. Green route for biomethane and hydrogen production via integration of biogas upgrading using pressure swing adsorption and steam-methane reforming process. *Renew. Energy* **2023**, *210*, 64–78. [CrossRef]

26. Chaemchuen, S.; Zhou, K.; Verpoort, F. From Biogas to Biofuel: Materials Used for Biogas Cleaning to Biomethane. *ChemBioEng Rev.* **2016**, *3*, 250–265. [CrossRef]

27. Ardolino, F.; Cardamone, G.F.; Parrillo, F.; Arena, U. Biogas-to-biomethane upgrading: A comparative review and assessment in a life cycle perspective. *Renew. Sustain. Energy Rev.* **2021**, *139*, 110588. [CrossRef]

28. Rostrup-Nielsen, J.; Christiansen, L.J. *Concepts in Syngas Manufacture*, 10th ed.; Catalytic Science Series; Imperial College Press: London, UK, 2011; ISBN 9781908978004.

29. LeValley, T.L.; Richard, A.R.; Fan, M. The progress in water gas shift and steam reforming hydrogen production technologies—A review. *Int. J. Hydrogen Energy* **2014**, *39*, 16983–17000. [CrossRef]

30. Full, J.; Ziehn, S.; Geller, M.; Miehe, R.; Sauer, A. Carbon-negative hydrogen production: Fundamentals for a techno-economic and environmental assessment of HyBECCS approaches. *GCB Bioenergy* **2022**, *14*, 597–619. [CrossRef]

31. Boot-Handford, M.E.; Abanades, J.C.; Anthony, E.J.; Blunt, M.J.; Brandani, S.; Mac Dowell, N.; Fernández, J.R.; Ferrari, M.-C.; Gross, R.; Hallett, J.P.; et al. Carbon capture and storage update. *Energy Environ. Sci.* **2014**, *7*, 130–189. [CrossRef]

32. Verduyn, M.; Geerlings, H.; van Mossel, G.; Vijayakumari, S. Review of the various CO_2 mineralization product forms. *Energy Procedia* **2011**, *4*, 2885–2892. [CrossRef]

33. Da Pires Mata Costa, L.; Micheline Vaz de Miranda, D.; Couto de Oliveira, A.C.; Falcon, L.; Stella Silva Pimenta, M.; Guilherme Bessa, I.; Juarez Wouters, S.; Andrade, M.H.S.; Pinto, J.C. Capture and Reuse of Carbon Dioxide (CO_2) for a Plastics Circular Economy: A Review. *Processes* **2021**, *9*, 759. [CrossRef]

34. Ravikumar, D.; Keoleian, G.A.; Miller, S.A.; Sick, V. Assessing the Relative Climate Impact of Carbon Utilization for Concrete, Chemical, and Mineral Production. *Environ. Sci. Technol.* **2021**, *55*, 12019–12031. [CrossRef]

35. Full, J.; Baumgarten, Y.; Dokur, Y.; Miehe, R.; Sauer, A. Biogas Plants as Hydrogen Production Facilities and Greenhouse Gas Sinks: Technology Comparison, Challenges and Potentials for Carbon Negative Hydrogen Production (HyBECCS). *Procedia CIRP* **2022**, *107*, 185–190. [CrossRef]

36. Tomita, E.; Kawahara, N.; Azimov, U. *Biogas Combustion Engines for Green Energy Generation*; Springer International Publishing: Cham, Swizterland, 2022; ISBN 9783030945381.

37. Mishra, A.; Kumar, M.; Bolan, N.S.; Kapley, A.; Kumar, R.; Singh, L. Multidimensional approaches of biogas production and up-gradation: Opportunities and challenges. *Bioresour. Technol.* **2021**, *338*, 125514. [CrossRef] [PubMed]

38. Sun, Q.; Li, H.; Yan, J.; Liu, L.; Yu, Z.; Yu, X. Selection of appropriate biogas upgrading technology-a review of biogas cleaning, upgrading and utilisation. *Renew. Sustain. Energy Rev.* **2015**, *51*, 521–532. [CrossRef]

39. Mallon, W.; Buit, L.; van Wingerden, J.; Lemmens, H.; Eldrup, N.H. Costs of CO_2 Transportation Infrastructures. *Energy Procedia* **2013**, *37*, 2969–2980. [CrossRef]

40. Becattini, V.; Gabrielli, P.; Antonini, C.; Campos, J.; Acquilino, A.; Sansavini, G.; Mazzotti, M. Carbon dioxide capture, transport and storage supply chains: Optimal economic and environmental performance of infrastructure rollout. *Int. J. Greenh. Gas Control* **2022**, *117*, 103635. [CrossRef]

41. Schaffelhofer, K. Neuer Gasmotor MAN E3872 mit 44.0% Wirkungsgrad und 735 kW Leistung aus nur 12 Zylindern, München. 2021. Available online: https://press.mantruckandbus.com/corporate/de/neuer-gasmotor-man-e3872-mit-44 0{-}{-}wirkungsgrad-und-735-kw-leistung-aus-nur-12-zylindern/ (accessed on 2 May 2023).

42. Mayr, K.; Hofer, F.; Ragowsky, G.; Gruber, W.; Arnberge, A.; Kabza, A.; Wolf, P.; Schmidt, M.; Jörissen, L. Systemvergleich Zwischen Wasserstoffverbrennungsmotor und Brennstoffzelle im Schweren Nutzfahrzeug: Eine Technische und Ökonomische Analyse Zweier Antriebskonzepte, Stuttgart. 2021. Available online: https://www.e-mobilbw.de/fileadmin/media/e-mobilbw/ Publikationen/Studien/e-mobilBW-Studie_H2-Systemvergleich.pdf (accessed on 2 May 2023).

43. Cullen, D.A.; Neyerlin, K.C.; Ahluwalia, R.K.; Mukundan, R.; More, K.L.; Borup, R.L.; Weber, A.Z.; Myers, D.J.; Kusoglu, A. New roads and challenges for fuel cells in heavy-duty transportation. *Nat. Energy* **2021**, *6*, 462–474. [CrossRef]

44. Schmidt, T. *Wasserstofftechnik: Grundlagen, Systeme, Anwendung, Wirtschaft*, 2nd ed.; Carl Hanser Verlag: München, Germany, 2022; ISBN 9783446473539.

45. Berne, E.K.; Berner, R.A. *Global Environment: Water, Air, and Geochemical Cycles*; Princeton University Press: Princeton, NJ, USA, 2012; ISBN 9780691136783.

46. Deutsches Institut für Normung e.V. DIN EN ISO, 14040:2021-02. Environmental Management—Life Cycle Assessment—Principles and Framework (ISO 14040:2006 + Amd 1:2020). German Version EN ISO 14040:2006 + A1:2020; Beuth Verlag: Berlin, Germany, 2021. [CrossRef]

47. Klöpffer, W.; Grahl, B. *Ökobilanz (LCA): Ein Leitfaden für Ausbildung und Beruf*; Wiley-VCH: Weinheim, Germany, 2009; ISBN 9783527320431.

48. Gupta, R.B. *Hydrogen Fuel: Production, Transport, and Storage*, 1st ed.; CRC Press: Boca Raton, FL, USA, 2008; ISBN 9781420045758.

49. Acar, C.; Dincer, I. Analysis and assessment of a continuous-type hybrid photoelectrochemical system for hydrogen production. *Int. J. Hydrogen Energy* **2014**, *39*, 15362–15372. [CrossRef]

50. Green, M.A. Hydrogen Safety Issues Compared to Safety Issues with Methane and Propane. *AIP Conf. Proc.* **2006**, *823*, 319–326. [CrossRef]

51. Moradi, R.; Groth, K.M. Hydrogen storage and delivery: Review of the state of the art technologies and risk and reliability analysis. *Int. J. Hydrogen Energy* **2019**, *44*, 12254–12269. [CrossRef]

52. Ullah Khan, I.; Hafiz Dzarfan Othman, M.; Hashim, H.; Matsuura, T.; Ismail, A.F.; Rezaei-DashtArzhandi, M.; Wan Azelee, I. Biogas as a renewable energy fuel—A review of biogas upgrading, utilisation and storage. *Energy Convers. Manag.* **2017**, *150*, 277–294. [CrossRef]

53. Mertins, A.; Heiker, M.; Rosenberger, S.; Wawer, T. Competition in the conversion of the gas grid: Is the future of biogas biomethane or hydrogen? *Int. J. Hydrogen Energy* 2023, in press. [CrossRef]

54. Kumar, S.; Kwon, H.-T.; Choi, K.-H.; Lim, W.; Cho, J.H.; Tak, K.; Moon, I. LNG: An eco-friendly cryogenic fuel for sustainable development. *Appl. Energy* **2011**, *88*, 4264–4273. [CrossRef]

55. Economides, M.J.; Sun, K.; Subero, G. Compressed Natural Gas (CNG): An Alternative to Liquefied Natural Gas (LNG). *SPE Prod. Oper.* **2006**, *21*, 318–324. [CrossRef]

56. Smajla, I.; Karasalihović Sedlar, D.; Drljača, B.; Jukić, L. Fuel Switch to LNG in Heavy Truck Traffic. *Energies* **2019**, *12*, 515. [CrossRef]

57. Gustafsson, M.; Svensson, N. Cleaner heavy transports—Environmental and economic analysis of liquefied natural gas and biomethane. *J. Clean. Prod.* **2021**, *278*, 123535. [CrossRef]

58. Ogden, J.; Jaffe, A.M.; Scheitrum, D.; McDonald, Z.; Miller, M. Natural gas as a bridge to hydrogen transportation fuel: Insights from the literature. *Energy Policy* **2018**, *115*, 317–329. [CrossRef]

59. Johansson, J.; Leirnes, J.; Walton, K.; Yilmaz, E.; Zindel, E. Hydrogen Power and Heat with Siemens Energy Gas Turbines: Reliable and Flexible Carbon-Free Energy, Erlangen. 2022. Available online: https://www.siemens-energy.com/global/en/offerings/ technical-papers/download-hydrogen-gas-turbine-readiness-white-paper.html (accessed on 3 May 2023).

60. Stephan, P.; Stephan, K. *Dubbel Taschenbuch für den Maschinenbau 1: Grundlagen und Tabellen*; Springer: Berlin/Heidelberg, Germany, 2020; ISBN 9783662597118.

61. Hermeling, W. *Handbuch für den LNG- und CNG-Praktiker*; Springer Fachmedien Wiesbaden: Wiesbaden, Germany, 2020; ISBN 9783658285517.

62. Bossel, U. Wasserstoff löst keine Energieprobleme. *TATuP* **2006**, *15*, 27–33. [CrossRef]

63. Trávníček, P.; Vítěz, T.; Koutný, T. The Equation of State of Biogas. *Acta Univ. Agric. Silvic. Mendel. Brun.* **2017**, *65*, 537–543. [CrossRef]

64. Kapoor, R.; Ghosh, P.; Kumar, M.; Vijay, V.K. Evaluation of biogas upgrading technologies and future perspectives: A review. *Environ. Sci. Pollut. Res. Int.* **2019**, *26*, 11631–11661. [CrossRef]

65. Liu, K.; Song, C.; Subramani, V. *Hydrogen and Syngas Production and Purification Technologies*; John Wiley & Sons Inc.: Hoboken, NJ, USA, 2010; ISBN 9780470561256.

66. Barton, J.W.; Bowers, S.L.; Butcher, T.G.; Harshman, R.A.; Lee, G.D.; Warfield, L. Appendix. In *Specifications, Tolerances, and Other Technical Requirements for Weighing and Measuring Devices*; National Institute of Standards and Technology: Gaithersburg, MD, USA, 2021. [CrossRef]

67. Apostolou, D.; Xydis, G. A literature review on hydrogen refuelling stations and infrastructure. Current status and future prospects. *Renew. Sustain. Energy Rev.* **2019**, *113*, 109292. [CrossRef]

68. European Commission. Fuel Price Comparison. 2022. Available online: https://alternative-fuels-observatory.ec.europa.eu/consumer-portal/fuel-price-comparison (accessed on 1 June 2023).

69. Sandhu, G.S.; Frey, H.C.; Bartelt-Hunt, S.; Jones, E. Real-world activity, fuel use, and emissions of heavy-duty compressed natural gas refuse trucks. *Sci. Total Environ.* **2021**, *761*, 143323. [CrossRef] [PubMed]

70. European Commission. Quarterly Report: On European Gas Markets. 2022. Available online: https://energy.ec.europa.eu/document/download/0e69ba99-07e9-4aed-8c34-dbfa3ae0add7_en?filename=Quarterly%20report%20on%20European%20gas%20markets%20Q1_2022.pdf (accessed on 19 June 2023).

71. Duan, Z.; Mei, N.; Feng, L.; Yu, S.; Jiang, Z.; Chen, D.; Xu, X.; Hong, J. Research on Hydrogen Consumption and Driving Range of Hydrogen Fuel Cell Vehicle under the CLTC-P Condition. *WEVJ* **2022**, *13*, 9. [CrossRef]

72. Abbas, H.F.; Wan Daud, W. Hydrogen production by methane decomposition: A review. *Int. J. Hydrogen Energy* **2010**, *35*, 1160–1190. [CrossRef]

73. Miyagawa, T.; Goto, M. Hydrogen Production Cost Forecasts since the 1970s and Implications for Technological Development. *Energies* **2022**, *15*, 4375. [CrossRef]

74. Poralla, M.; Honegger, M.; Ahonen, H.-M.; Michaelowa, A.; Weber, A.-K. *Sewage Treatment for the Skies: Mobilising Carbon Dioxide Removal through Public Policies and Private Financing*; NET-Rapido Consortium and Perspectives Climate Research: London, UK; Freiburg im Breisgau, Germany, 2021.

75. Puro.Earth. CORC Supplier Listing. Available online: https://puro.earth/CORC-CO2-removal-certificate (accessed on 15 May 2023).

76. European Commission. Certification of Carbon Removals—EU Rules. Available online: https://ec.europa.eu/info/law/better-regulation/have-your-say/initiatives/13172-Certification-of-carbon-removals-EU-rules_en (accessed on 1 June 2023).