



Article

A New Perspective for Climate Change Mitigation—Introducing Carbon-Negative Hydrogen Production from Biomass with Carbon Capture and Storage (HyBECCS)

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Abstract: The greatest lever for advancing climate adaptation and mitigation is the defossilization of energy systems. A key opportunity to replace fossil fuels across sectors is the use of renewable hydrogen. In this context, the main political and social push is currently on climate neutral hydrogen (H₂) production through electrolysis using renewable electricity. Another climate neutral possibility that has recently gained importance is biohydrogen production from biogenic residual and waste materials. This paper introduces for the first time a novel concept for the production of hydrogen with net negative emissions. The derived concept combines biohydrogen production using biotechnological or thermochemical processes with carbon dioxide (CO₂) capture and storage. Various process combinations referred to this basic approach are defined as HyBECCS (Hydrogen Bioenergy with Carbon Capture and Storage) and described in this paper. The technical principles and resulting advantages of the novel concept are systematically derived and compared with other Negative Emission Technologies (NET). These include the high concentration and purity of the CO₂ to be captured compared to Direct Air Carbon Capture (DAC) and Post-combustion Carbon Capture (PCC) as well as the emission-free use of hydrogen resulting in a higher possible CO₂ capture rate compared to hydrocarbon-based biofuels generated with Bioenergy with Carbon Capture and Storage (BECCS) technologies. Further, the role of carbon-negative hydrogen in future energy systems is analyzed, taking into account key societal and technological drivers against the background of climate adaptation and mitigation. For this purpose, taking the example of the Federal Republic of Germany, the ecological impacts are estimated, and an economic assessment is made. For the production and use of carbon-negative hydrogen, a saving potential of 8.49–17.06 MtCO₂,eq/a is estimated for the year 2030 in Germany. The production costs for carbon-negative hydrogen would have to be below 4.30 € per kg in a worst-case scenario and below 10.44 € in a best-case scenario in order to be competitive in Germany, taking into account hydrogen market forecasts.

Keywords: negative emission technologies; carbon dioxide removal; HyBECCS; hydrogen bioenergy with carbon capture and storage; biohydrogen; carbon-negative hydrogen; climate positive hydrogen; CCS; BECCS; DACCS; NET; CDR



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1. Introduction

The Paris Accord was adopted in 2015 to significantly limit the risks and impacts of climate change. The participating nations committed themselves to keeping the increase in the global mean temperature well below 2 K compared to pre-industrial levels and to making efforts to limit the increase even to 1.5 K. To achieve these agreements, measures are to be taken that will lead to a balance of greenhouse gas (GHG) emissions and sinks, i.e., greenhouse gas neutrality, in the second half of the 21st century [1]. Therefore, laws and policies have been introduced worldwide. For example, the European Union (EU)

resolved the European Green Deal in 2019, which includes several key measures to reach greenhouse gas neutrality by 2050 within the EU [2]. Numerous resolutions and legislations were also passed at national levels. The Federal Republic of Germany, for example, has set the Climate Protection Program 2030 and the Climate Protection Act, which came into force in December 2019 [3,4].

Burning fossil fuels for gaining energy has the greatest impact on climate change in industrialized nations. Energy-related emissions account for about 81% of greenhouse gas emissions across Europe [5] and about 85% in Germany [6]. The steady expansion of renewable energy is therefore considered a key lever for reducing greenhouse gas emissions. These efforts currently take into account mainly climate-neutral electrical energy by wind power and solar plants [2,7]. However, some of the greenhouse gas emissions cannot be avoided through electrification and expansion of renewable electricity generation. For example, sectors such as the steel, chemical, and cement industries are particularly difficult to defossilize, because electrification is not sufficient [8–10]. Moreover, in the transport and mobility sectors, complete electrification is hardly feasible, especially for large commercial vehicles or in aviation and shipping, due to the limited gravimetric energy density of currently existing battery technologies [11–15]. Renewable chemical energy carriers with high energy densities, generated either via electrochemical, so-called power-to-x processes or from renewable biomass, are better suited for this purpose [7,16]. In this context, sustainably produced green hydrogen is seen as a key chemical energy carrier of the future [7,10,17,18]. Green hydrogen can be produced through electrolysis using renewable electricity or from renewable biomass, either through thermochemical processes or by using microorganisms and biotechnological methods. Hydrogen produced from biomass is also referred to as biohydrogen and can lead to a more efficient use of biogenic raw materials exploiting residual and waste materials.

However, even with a complete conversion of the electrical energy supply to renewable power generation and a successful defossilization of the heat and transport sectors, residual emissions remain in areas such as agriculture and waste management. These emissions must be compensated to net zero GHG emissions by 2050 [7]. GHG emissions in the earth's atmosphere are therefore to be actively reduced using negative emission technologies (NET). The latest special report by the Intergovernmental Panel on Climate Change (IPCC) outlines development scenarios that could limit global warming to less than 1.5 K. In all success scenarios, NET are essential components that will play an increasingly important role in climate adaptation and mitigation by 2050 [19]. Known NET include technologies that achieve a negative carbon footprint by capturing the greenhouse gas CO₂ and permanently storing (CCS—Carbon Capture and Storage) or using (CCU—Carbon Capture and Utilization) it [19]. The most relevant NETs are the direct capture of CO₂ from ambient air (DACCS) and Bioenergy with Carbon Capture and Storage (BECCS), where energy sources are extracted from the biomass and the resulting co-product CO₂ is captured [19,20].

This paper introduces a new NET concept combining biohydrogen production with carbon capture and storage, referred to as HyBECCS. HyBECCS couples the sustainable production of hydrogen from renewable biomass, for example from residual and waste streams, with efficient CO₂ capture and subsequent storage. The hydrogen produced has a negative carbon footprint and is therefore referred to as carbon-negative hydrogen or climate-positive hydrogen. The technological advantages of this new concept can support climate adaptation and mitigation in the future by approaching the GHG emission problem from two sides. On the one hand, it contributes to the defossilization, bringing more emission-free hydrogen into use, especially for sectors that are difficult to electrify, and, on the other hand, it contributes to the compensation of the remaining residual emissions through the active CO₂ extraction from the atmosphere. The availability and suitability of organic feedstocks, microorganisms, and process technologies, as well as the economic relations and ecological impacts of the HyBECCS approach need to be investigated to be able to solve societal and environmental problems. This paper gives the basics for

this purpose through defining the new approach and describing related technologies, discussing technological properties, and assessing ecological and economic benefits for climate adaptation and mitigation using the example of Germany.

2. Objectives and Enablers

The HyBECCS concept describes technology combinations that merge the production of hydrogen from renewable biomass with subsequent carbon dioxide capture and storage or use. The main objectives of the HyBECCS concept introduced in this section are given by climate adaptation and mitigation goals. Hydrogen has a key role to play in this context, as it can greatly contribute to the defossilization of critical industrial areas that are difficult to electrify, such as steel production, chemical industry, cement production, or heavy-duty transport industry. Therefore, many countries have already adopted national strategies to establish sustainably produced green hydrogen as a basic energy carrier. These include, for example, the US, Canada, Japan, France, Norway, the Netherlands, Australia, and Chile [21–28]. The use of carbon-negative hydrogen produced with HyBECCS concepts can additionally compensate for residual emissions by achieving a reduction of the GHG concentration in the atmosphere through active CO₂ capture and storage. For companies, the use of carbon-negative hydrogen offers an opportunity to improve their carbon footprint. Its use can have a positive impact on their GHG emission balances and can make a direct and effective contribution to climate change mitigation. Besides, HyBECCS can contribute to a more efficient use of existing raw materials by residual and waste exploitation. However, to provide sustainable value creation, carbon-negative hydrogen must be competitive and have undergone a holistic ecological analysis to ensure positive environmental impacts. The HyBECCS concept must therefore be ecologically as well as economically analyzed and optimized. Therefore, the most important basis is a technical understanding of the underlying processes, the fundamentals of which are explained in the following sections.

Furthermore, societal, political, and scientific developments that could encourage the dissemination of carbon-negative hydrogen and the application of HyBECCS technologies can be observed. One societal factor that has gained momentum in science and politics in recent years is the so-called biological transformation of the industry and society toward a biointelligent value creation which is closely interlinked with the global efforts of building a sustainable bioeconomy [29–31]. The process of biological transformation describes an increasing use of biology and its materials, principles, and organisms to create new and sustainable value creation systems. This development is enabled by the increasing interconnection of biotechnology, production technology, and information technology. The creation of a sustainable energy supply system is a major field of action and objective of this transformation process, which can lead to a sustainable bioeconomy in the form of a technology-based subsistence economy [32,33]. Key characteristics of these activities are efficient and coupled material and energy carrier cycles as well as utilization cascades which result in energy recovery [29,31,34,35]. Accordingly, the introduced HyBECCS concept can be seen as part of the biological transformation and bioeconomy developments and is likely to be strengthened from the momentum they generate.

3. State of the Art

This section introduces hydrogen usage as an energy carrier and raw material and its particular importance for achieving climate protection targets (Section 3.1). A special focus is placed on the sustainable production of biohydrogen from biomass, as it is the key sub-process for the HyBECCS approach presented (Section 3.2). Further, existing technology approaches for NET are explained, and the respective advantages and disadvantages are discussed (Section 3.3).

3.1. The Future of Hydrogen

Hydrogen as an energy carrier has many advantages and will play an important role in the energy infrastructure and circular economy of the future [36,37]. Since hydrogen can be produced in a climate-neutral manner, and only water is released during its use, it is considered a climate-friendly energy carrier [36–39]. Hydrogen that is produced from renewable sources is generally referred to as “green hydrogen”. It is contrasted with the so-called “grey hydrogen”, which is referred to as hydrogen produced from fossil, non-renewable sources [18]. Currently, most of the global hydrogen production consists in grey hydrogen produced by steam reforming of fossil natural gas [8,37]. Grey hydrogen can be made carbon-neutral by applying carbon capture and storage (CCS) technology and is then called “blue hydrogen”, but it stays a non-renewable source [18,40]. We introduced “carbon-negative hydrogen” to distinguish between carbon-neutral green or blue hydrogen. Due to the high gravimetric energy density and good storage and transportability, there are many possible applications for hydrogen [37]. Fuel cells can be used to generate electric power as well as heat from hydrogen. Hydrogen can also be used as a reducing agent in the steel industry [8]. It is considered an inevitable basic building block in sectors that are difficult to electrify [37].

In addition to electrolysis using renewable electricity, green hydrogen can be produced from biomass, e.g., from organic residual and waste streams referred to as biohydrogen. A distinction is made between thermochemical and various biotechnological generation pathways for biohydrogen, which will be discussed in the following.

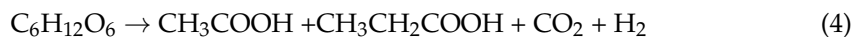
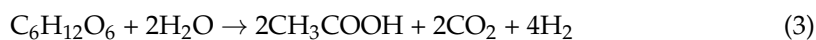
3.2. Biohydrogen Production

Biohydrogen production technologies can be divided into thermochemical and biotechnological approaches. On an industrial scale, biohydrogen can be produced from biomass (wood, straw, grass cuttings, etc.) but also from other bioenergy sources (biogas, bioethanol, etc.) using thermochemical processes. Thermochemical processes can be subdivided into gasification and pyrolysis. Depending on the feedstock used, the synthesis gas formed during gasification or pyrolysis gas consists of varying proportions of hydrogen (H₂), carbon dioxide (CO₂), carbon monoxide (CO), methane (CH₄), and other components such as nitrogen, water vapor, light hydrocarbons, hydrochloric acid, alkali chlorides, sulfur compounds, biochar, or tar [41,42]. Using subsequent steam reforming and/or water gas shift reactions, the following chemical reactions take place, resulting in the production of CO₂ and hydrogen [43,44].



Biotechnological biohydrogen production processes using suitable microorganisms in adequate production facilities can be divided into three main categories: biophotolysis, photofermentation, and dark fermentation [45,46]. Dark fermentation is particularly promising because it is easier to scale up without exposure due to its simpler structure [47–50]. Moreover, technological maturity is the most advanced among the biotechnical processes and thus also relevant knowledge, for example about process parameters such as product gas compositions, is known. Therefore, in this paper, a special focus is given to dark fermentation, knowing that there are alternative processes or process combinations with even higher potential. These include, for example, combinations, e.g., of dark fermentation and photofermentation [51–53]. Sugars and starchy products are particularly suitable feedstocks for dark fermentation [48]. However, with pretreatment, a variety of feedstocks such as food waste, wood, or wastewater can be used for biological hydrogen production [46,49,54,55]. In dark fermentation, the contained glucose is decomposed into hydrogen, carbon dioxide, and the byproducts acetic acid (3), propionic acid (4), or butyric acid (5) [39,48,49,56,57]. The

following reaction equations form the basis of these syntheses and take place enzymatically catalyzed via metabolic processes of the microorganisms used.



The net energy ratio (NER) of biohydrogen production of 1.9 is significantly better than conventional hydrogen production with methane steam reforming, which has a NER of 0.64 [58]. Utilizing the resulting byproducts in one- or two-stage processes [48,56,58] can further improve the NER. For example, coupling dark fermentation with photofermentation can further decompose the acetic acid components producing hydrogen and increase the NER to 3.1 [58]. Both single organisms and mixed bacterial cultures can produce other byproducts in a biohydrogen plant such as acetone, ethanol, or butanol [48]. Furthermore, adding nanoparticles of various metal oxides can also result in a better hydrogen yield [56]. Thus, further improvements of the processes can be expected through continuous research [39,49–51,53,56]. The product gas of biohydrogen production consists of 30–65% H₂ and 70–35% CO₂, depending on the process parameters, such as the type of microorganism, temperature and pH [52,59–65]. Hydrogen sulfide, water, and nitrogen are also possible by-products [66,67].

3.3. Negative Emission Technologies

NET create greenhouse gas sinks through anthropogenic activities. Sinks that are not directly influenced by humans are not included [19]. Most prominent NET are also known CCS technologies which include, for example, BECCS and DACCS. They can be classified as NET if CO₂ is removed from the atmosphere and negative emissions are generated. However, if the carbon comes from fossil sources, the net emissions are at best zero. In this case, it cannot be classified as NET. In general, CCS technologies can be divided into two steps: first, the capture and sequestration; and second, the storage of CO₂. To capture and sequester CO₂ from gas mixtures in the first step, technologies are needed that separate CO₂ from gas mixtures. Four approaches (absorption, adsorption, cryogenic separation, and membrane separation) have already been tested and are used for CCS technologies [68–76]. The differences between those options in their effectiveness, robustness, energy requirements, and costs regarding their potential use for HyBECCS concepts are discussed below.

Absorption uses a solvent such as water or polyethylene glycol in which CO₂ is first dissolved and then released by changing the conditions [68]. Pressurized water scrubbing, for example, is used in biogas plants to separate CO₂ and H₂S from methane. In this process, water serves as an absorber for CO₂ through increased pressure. This absorber technology is considered comparatively robust and cost-effective [69,70]. Adsorption, in contrast, occurs at the surface of solids [76]. Again, CO₂ remains on the solid sorption material under certain conditions and can be regenerated by a change in temperature, pressure, or humidity [74]. A technically mature adsorption process is pressure swing adsorption on activated carbon. At a low temperature, of about 5 °C, and increased pressure, of about 2–7 bar, CO₂ accumulates on the surface of the activated carbon. The gas must first be free from impurities such as sulfur or dust for the activated carbon to achieve long lifetimes of up to 20 years [72]. Cryogenic separation takes advantage of the different condensation temperatures of the gases. For example, CO₂ condenses at −78.5 °C and hydrogen at −252 °C [77]. At very low temperatures the liquified CO₂ can easily be separated from other components still in a gaseous state. A very high purity of the recovered CO₂ fraction of 99.9% can be achieved, but this requires a comparatively high energy input. Membrane technology, in which the selective permeability of gases through certain material is exploited, is particularly suitable for gas mixtures that already contain a very high CO₂ content, of more than 90% [72–74]. Membrane technologies

are considered to be more environmentally friendly and cost-effective than the other options described [67,78]. This is due to the simpler modular design and lower energy requirements [76,78]. Challenges include selectivity, permeability, and sensitivity to sulfur compounds [68,74,79]. Further, the comparatively low purity of the CO₂ fraction after separation is to be mentioned. The difference in purity, however, moves in a wide range, from 80–99.9999%, and depends on the membrane technology chosen and the materials used [78,80,81].

In general, gas mixtures with a relatively low CO₂ concentration require technologies with higher selectivity to effectively separate CO₂ [82]. It is thermodynamically advantageous if the gas mixture to be separated already contains a large amount of CO₂ [83]. Thus, the CO₂ source has a strong influence on the choice of the separation process and the costs per captured tonnes of CO₂. In this context, the initial situations are differentiated between the separation of CO₂ from ambient air or from so-called point sources, which already have a significantly higher CO₂ concentration than the ambient air. The possibility to capture CO₂ from ambient air is known as Direct Air Capture (DAC). The obvious advantage of DAC is that the CO₂ concentration in the atmosphere can directly be lowered and does not require point sources [84,85]. However, because the ambient air has a relatively low CO₂ concentration, of about 400 ppm, on average, the capture of CO₂ is thermodynamically less favorable than using point sources [83]. This is a reason why BECCS outperform, e.g., DACCS in terms of the primary energy required per tonne of carbon sequestered [86]. Chemical absorbers or adsorbers that undergo a chemical reaction with CO₂ are often used for DAC [71,72]. Because of the strong binding and selectivity, even the small amount of CO₂ in the air mixture can be filtered out. However, more energy must be expended for the regeneration of the CO₂, and a large volume of air must be passed through the equipment to capture CO₂ in appreciable quantities. This results in high energy requirements and high costs for DAC [20,82,87]. If the high energy requirement for DAC is not carried out exclusively with renewable excess energy that does not compete for other uses, this can even lead to a positive emission balance instead of net negative emissions [20]. The second option is to capture CO₂ at point sources that already contain a higher concentration of CO₂. These include, for example, exhaust gases from coal or gas-fired power plants, bioenergy plants, or other industrial facilities. The higher CO₂ concentration of the gas mixture allows a higher energy efficiency of gas separation [87]. However, if the gas mixture has a complex and pollutant composition, the separation can be very costly and technologically complex [69]. Therefore, post-combustion carbon capture (PCC), in which combustion exhaust gases are filtered, is in many cases cost-ineffective, because the heterogenous gas composition is harmful to the separation technology [72]. Additionally, it cannot be classified as NET if the carbon originates from fossil sources, as in the case of PCC at coal-fired power plants.

The second step of CCS technology is the permanent storage of the captured CO₂. This can be done, for example, in geological formations such as aquifers [88]. Furthermore, there is the possibility to upgrade and use CO₂ for new products like plastics or lithium carbonate for accumulators. If the captured CO₂ is used, this is referred to as CCU technologies [89]. However, there is no consensus on whether this is a NET because of the limited lifetime of the products. It may only be a delay in emission [90]. But not only CO₂ can be used to achieve negative emissions. Residues from processed biomass can increase the carbon content of soil in so-called soil carbon sequestration and therefore lead to negative emissions [91,92]. Beyond the described CDR technologies, naturally occurring processes that can be influenced by humans, such as reforestation or enhanced weathering, also fall under the term NET. However, these are not relevant to the derivation of the introduced HyBECCS approach and are therefore not considered in this paper.

BECCS is highlighted in the IPCC Special Report as one of the key NET on achieving the 1.5 K target [19]. Additionally, the presented HyBECCS concept can be classified as a new BECCS technology approach. Therefore, BECCS technologies are discussed in more detail in the following paragraph.

With BECCS technologies, plant-based biomass is used for energy generation, and the CO₂ that is released can be captured and permanently removed from the atmosphere [93,94]. Regarding BECCS, three basic ways of generating energy from plant biomass are distinguished: the thermochemical conversion, the physicochemical conversion, and the biological conversion of biomass to solid, liquid, or gaseous energy sources [95]. One example of thermochemical conversion is incomplete combustion or pyrolysis. In this process, wood and stalk-type biomass are converted into coal, pyrolysis oil, and various gases [69]. The relative proportions of solids, liquid, and gas depend on the combustion parameters and residence time in the combustor [96]. Depending on the feedstock and process conditions, gas mixtures are produced that contain H₂, CO, CH₄, and CO₂ as well as water vapor, nitrogen, particles, tar, hydrochloric acid, alkali chlorides, and sulfur compounds, as described in Section 3.2 [97,98]. The gas mixture must then be separated and processed for use as fuel [69,95–100]. Physicochemical conversion can be used for energy recovery from plant-based oils. They can be mechanically pressed or extracted from plants and then be chemically upgraded, for example by transesterification [101]. The resulting so-called biodiesel is a well-known product from physicochemical conversion processes, but research is also being conducted on the production of kerosene substitutes for aviation from algae oil, for example [102,103]. In biological conversion, biomass is decomposed by microorganisms and converted to methane, hydrogen, or alcohol, for example. A well-known biological conversion process is agricultural biogas production [94]. In conventional biogas plants, microorganisms produce a mixture consisting of 40–75% methane and 15–60% CO₂. Impurities such as H₂S are other possible components that can occur [57,75]. To use it as biogas, this mixture is separated. Biogas with high methane contents can be transported better, and its calorific value increases [68,75]. The CO₂ can be captured and permanently removed from the atmosphere in storage facilities, thus causing negative emissions. Moreover, the digested residue can be used as fertilizer, potentially leading to a further increase of the negative emission effect [92]. However, when the methane biogas is used, CO₂ is released into the atmosphere unless another CCS measure is implemented.

4. Basic Considerations for the HyBECCS Concept

In this section, the HyBECCS concept is described in detail, deriving main technological advantages that lead to lower costs and energy efficiency compared to competing technologies, such as DACCS and other BECCS processes.

The HyBECCS concept summarizes modular process combinations that can be split into four basic process steps (cf. Figure 1): the biomass pretreatment, the biohydrogen production, the product gas separation, and its processing. The authors suggest drawing the system border for economic and ecological classification and balancing around this four steps. Using a consistent system border will make it easier to compare different HyBECCS process combinations. For each of the four steps there are variable sub-processes which have either already reached market maturity or whose functionality has been demonstrated and tested at laboratory scale or prototype scale. Many of them are described in Section 3. However, a HyBECCS process combination has never been experimentally implemented as a whole process linking all of the four steps. Respective technological peculiarities, challenges, and opportunities in all of the four steps are discussed below. The subsequent processes of transportation and storage or utilization of CO₂ and options of hydrogen usage are not in the focus of consideration in this paper, since they have already been discussed and compared in other publications [104–107]. In addition, biomass production and transportation is not included in this work. The following diagram outlines the modularity of the HyBECCS approach described in the following.

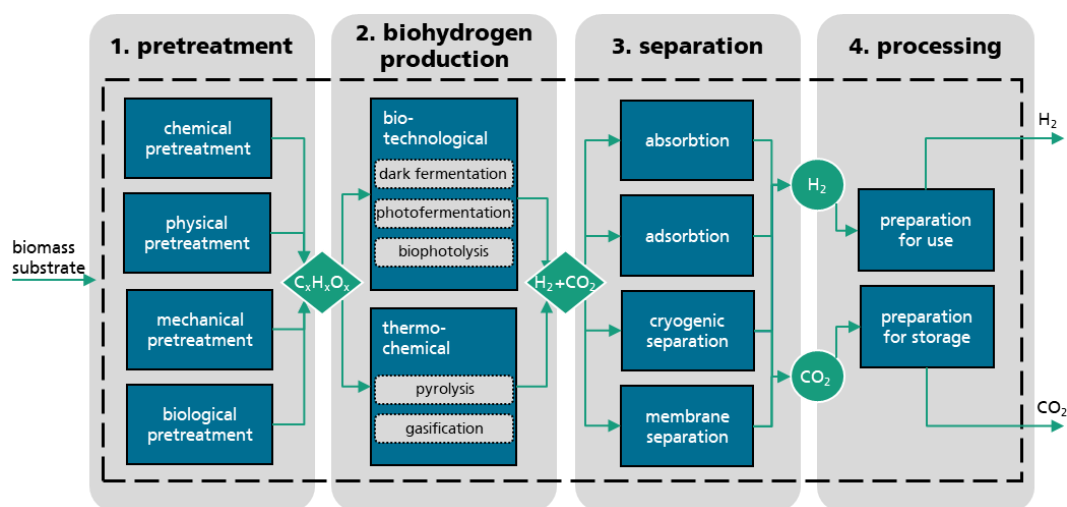


Figure 1. Process diagram for carbon-negative hydrogen production using the Hydrogen Bioenergy with Carbon Capture and Storage (HyBECCS) approach.

For the key step of all HyBECCS approaches to biohydrogen production, the possible feedstocks are very diverse, as described in Section 3.2. However, upstream pretreatment processes (step 1) are required in most cases. For example, to be able to apply biotechnological fermentation processes, residual and waste materials containing sugar are usually pre-filtered and woody biomasses have to undergo a saccharification process. Substrates containing starch, fats, or proteins are subjected to hydrolysis pretreatment releasing single sugars, amino acids, and fatty acids for further microbial metabolism, as is common, for example, in most existing biogas production plants [108–113]. In addition, mechanical comminution or physical digestion methods are necessary. They are also needed for many thermochemical biohydrogen production processes. Moreover, thermochemical processes often require preparatory drying. One challenge of the pretreatment step is therefore the efficient processing of the widest possible range of residual and waste materials for use in biohydrogen processes. For the biohydrogen production (step 2), there is a choice of different thermochemical or biotechnological process options or combinations, as described in Section 3.2, that must be selected according to the specific characteristics of the available biomass streams and pretreatment options. Relevant for subsequent steps and the overall efficiency of the HyBECCS approach is the product gas composition resulting from the biohydrogen production process. In most cases, hydrogen and carbon dioxide are the main outputs [52,59–65]. The proportions of these two components should be maximized to exploit the ecological and economic potential and prepare for efficient gas separation (step 3). The lower the heterogeneity of the gas mixture to be separated and the higher the proportions of the main fractions H_2 and CO_2 , the better the conditions for efficient gas separation. Using biotechnological biohydrogen production processes, for example, the mixture usually does not contain combustion residues or other harmful gases that can negatively affect the downstream gas separation, as is the case with PCC or some thermochemical biohydrogen production processes [87]. To fractionate the product hydrogen for use and the carbon dioxide for downstream storage or use, a suitable gas separation process which can be carried out in various options is to be chosen, as described in Section 3.1. To separate H_2 and CO_2 , the physical and chemical prerequisites of an efficient separation are given, for example, due to the different polarities and molecular sizes. Membrane technologies, for example, are an attractive alternative to conventional processes, such as pressure swing adsorption or cryogenic distillation, due to their comparatively high separation quality and energy efficiency [67]. As described in Section 3.1, the disadvantage of DAC regarding the gas separation step is the low content of CO_2 in the ambient air, leading to higher energy consumption and high plant investment compared to the point source resulting from, e.g., biohydrogen production processes [83]. In the final step of processing

(step 4), the separated CO₂ has to be compressed or otherwise prepared for storage or use, and hydrogen has to be compressed or liquefied and purified so that it can either be fed into the natural gas network or transported to users, e.g., in composite pressure tanks. The carbon-negative hydrogen produced can be used materially for chemical processes or by means of fuel cells as energy carriers to generate heat and electricity or to power vehicles, as described in Section 3.1. Using fuel cells, the reaction with oxygen from the ambient air produces only water. In contrast to hydrocarbon-based e-fuels or bio-fuels such as biogas, no greenhouse gas emissions are emitted during its use. This results in an inherently higher CO₂ capture potential for HyBECCS, leading to a decisive advantage over BECCS technologies that aim to produce hydrocarbons as energy carriers. However, to use hydrogen in fuel cells, purity requirements must be met, which are specified in European standards, for example. The purity of hydrogen for its use in hydrogen fuel cells is specified therein at the value 3.7, which corresponds to a purity of 99.97% [114]. The following three points summarize the key advantages of HyBECCS concerning their environmental and economic potential:

1. The product gas composition of biohydrogen processes consisting mainly of H₂ as the main product and CO₂ as a co-product makes the capturing of CO₂ energy- and cost-effective compared to low concentrated CO₂ sources like ambient air [52,59–65,83,87,115].
2. The possibility to use biotechnological methods for biohydrogen production within HyBECCS approaches leads to carbon sources that are not polluted with combustion residues. In comparison to PCC, there is no risk to impair the subsequent separation or further processing [66,67,69,87].
3. Hydrogen is a carbon-free energy carrier. Since there is no carbon remaining in the product, HyBECCS approaches have a comparatively high cumulative CO₂ capture rate and thus a larger specific GHG emission reduction potential compared to, e.g., BECCS technologies producing hydrocarbon-based biofuels [36–39].

Due to these advantages, the HyBECCS concept can be seen as a new promising perspective within BECCS methods. However, to be able to make conclusive statements on process ecology and economics, more in-depth techno-economic analyses and ecological assessments must be carried out. The following section gives an insight into these assessments using the example of Germany.

5. General Perspectives for HyBECCS Approaches

Due to the advantages described in Section 4, HyBECCS offers a perspective for sustainable and efficient production of hydrogen as well as climate adaption and mitigation. Associated political regulatory measures such as CO₂ pricing and emissions trading could result in monetary credits from which the HyBECCS concept could benefit. Further, the global political and societal push toward establishing green hydrogen as a sustainable basic energy carrier across several sectors will have a positive effect for carbon-negative hydrogen production by better meeting the resulting demand. Furthermore, the increasing efforts of companies to leverage its externalities through carbon footprint improvements for business purposes in so-called social or green marketing concepts are also potential drivers of the HyBECCS concept [7,10,18,116,117]. An important basis for further development and industrial application of HyBECCS is a fundamental understanding of their economic and ecological impacts. In the following, from the example of Germany, possible impacts for climate adaptation and mitigation (Section 5.1) and the economic viability of HyBECCS concepts are discussed (Section 5.2).

5.1. Ecological Classification

The amount of sustainably available biomass is limited and should therefore be used as efficiently as possible. One promising way to do so is carbon-negative hydrogen production. The main ecological advantage of this possibility in terms of climate adaption and mitigation is the removal of CO₂ from the atmosphere. Compared to other NET, this can be done particularly efficiently by HyBECCS, as described in Section 4. In the

following, an estimate of the GHG emission reduction potential of HyBECCS in Germany is carried out, starting from bioenergy potentials already investigated for biohydrogen production through dark fermentation processes. Biohydrogen production as a stand-alone technology is already considered as a potentially climate-neutral process with a wide substrate spectrum. Among the options described in Section 3.2, biotechnological pathways allow for hydrogen production with less energy-consumption than thermochemical and electrochemical processes, which is due to lower process temperatures and pressures [46]. Besides, biotechnological hydrogen production has the advantage of offering a carbon source with high purity and no combustion residues, that could impair the subsequent separation or purification of CO₂, as described in Section 4. Thermochemical processes like pyrolysis or gasification are disadvantaged in these points. In the following, a special focus will therefore be placed on a biotechnological hydrogen production process as the basis for HyBECCS. In this investigation, the best-known and most widespread biotechnological approach, dark fermentation, is considered for the following ecological assessment. Further, the analysis is reduced to the environmental impact category global warming and to Germany as the spatial and systemic frame of reference.

The future potential of biohydrogen production from residuals through dark fermentation has been investigated in a recent study for Germany [118]. It should be noted that only biomass potentials from residues were taken into account in this consideration, which serves as a basis for the subsequent analysis. Social conflicts concerning land-use, for example through competitive situations with the production of food and feed, are thus not to be expected. For Germany, it was estimated that a theoretically achievable annual energy supply of 9.9 TWh/a up to 19.9 TWh/a will be possible in 2030 through dark fermentation of residual and waste materials. This is the same order of magnitude as that of Germany's targets for the production of hydrogen by electrolysis using renewable electricity. Within the national borders, these are set up to 14 TWh/a until 2030 [18]. Assuming that this energy amount of green hydrogen from dark fermentation substitutes an equal amount of energy provided by natural gas, this results in a saving of 2.0–4.0 MtCO₂,eq/a. In addition to natural gas, coal, and lignite, grey hydrogen or heating oil could also be substituted, which would lead to even greater savings due to higher specific GHG emissions related to the amount of energy produced [119]. Besides, the substitution of electricity from fossil sources is possible using hydrogen fuel cells. However, the replacement of natural gas serves as a plausible assumption, since hydrogen can be used directly in the mixture with natural gas in many cases without technological adjustments and can also be fed directly into natural gas distribution networks [18]. HyBECCS approaches further increase the GHG savings potential by creating negative emissions, as described in Section 4. Assuming that the entire amount of CO₂ produced as a by-product during dark fermentation processes were to be separated and stored, 6.4–13.0 MtCO₂,eq/a of negative GHG emissions could be achieved in Germany by 2030. This assumption was based on a realistic stoichiometric-based product gas composition of fermentative biohydrogen production through the dark fermentation of CO₂ and H₂ in equal quantities, as can also be seen in Section 3.2 and Equations (4) and (5) [64–70]. In total, the use of carbon-negative hydrogen, under the assumptions made, results in a saving potential of 8.49–17.06 MtCO₂,eq/a due to CO₂ avoidance through fossil energy source substitution and its active removal from the atmosphere. In Figure 2, the estimated emission reduction potentials using HyBECCS are summarized.

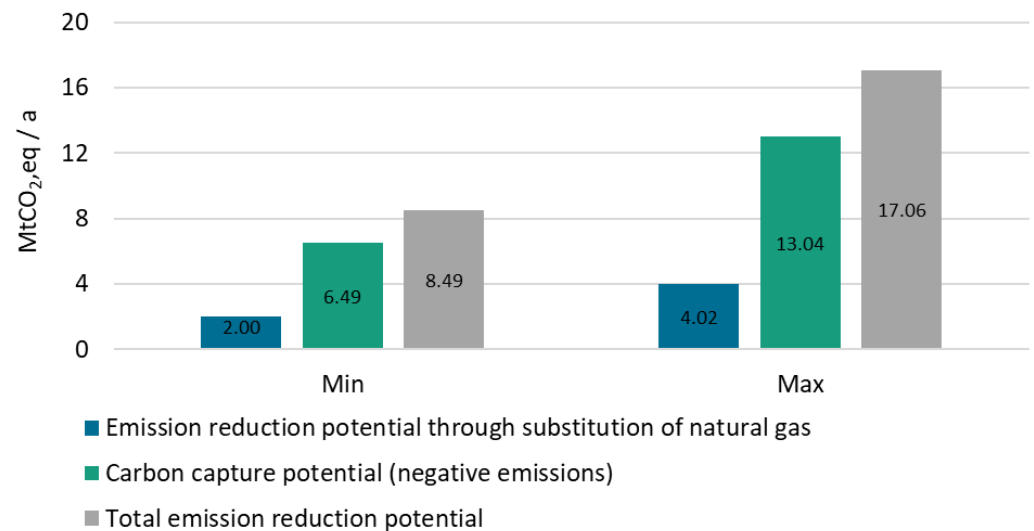


Figure 2. Emission reduction potential through HyBECCS in Germany based on carbon-negative hydrogen production from residual biomass using dark fermentation.

The most important challenge in tackling climate change at a global level is the reduction of GHG emissions. However, a sole consideration of the GHG balance is not sufficient to explore the sustainability of HyBECCS approaches [115,120]. Further impact categories within life cycle assessments (LCA) according to DIN EN ISO 14040/44 are to be considered. Based on EEA classifications 6 and 10 for the chemical industry and agricultural products according to Grosse-Sommer et al., this also includes resource depletion, acidification, consumptive water use, eutrophication, human toxicity potential, photochemical ozone formation, ozone depletion, and land use [121]. Furthermore, the impact categories biodiversity loss and ecosystem services should also be considered [122]. However, this paper cannot make a final assessment taking into account all relevant impact categories. For a holistic view and final assessment, further investigations are still needed.

5.2. Economic Classification

The use of carbon-negative hydrogen can be economically attractive for companies, as it can provide a way to improve their corporate image, reducing the company's carbon footprints, and contribute to social or green marketing concepts. However, to be broadly applied, carbon-negative hydrogen must also be price-competitive, and its production must be profitable. The profitability depends on many internal and external impact factors, some of which are summarized in Figure 3. The internal impact factors describe influences that emanate directly from the technical setup and equipment used for the HyBECCS process combination as described in Section 4, leading to differences in CO₂ separation rates, hydrogen yield, energy efficiency, or plant investment costs, for example. Internal factors are necessary information for comprehensive techno-economic analyses. However, they are very difficult to estimate for HyBECCS due to the low level of technological maturity of the central process step of biohydrogen production and the lack of corresponding industrial-scale production technologies as data sources. Although further research in this regard has already been initiated by the authors, the information situation is not yet sufficient for a comprehensive analysis.

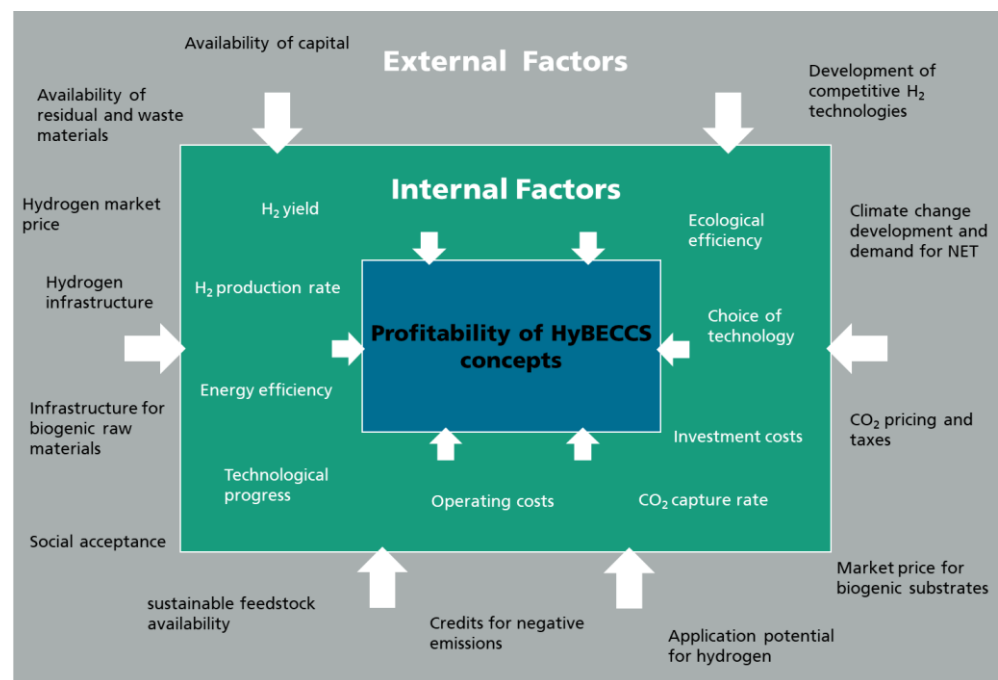


Figure 3. Impact factors on the profitability of carbon-negative hydrogen and HyBECCS.

External factors, in contrast, include forces that affect the competitiveness of carbon-negative hydrogen in the marketplace and are not directly dependent on the technical setup of the chosen process combination and technical setup. These include, for example, market or policy conditions affecting the price of hydrogen or the profitability of carbon-negative hydrogen production through financial incentives. A key external factor in this context are political regulatory measures from environmental and climate policies. Thereby, for the political regulation of GHG emissions via financial incentive systems, a distinction can be made between the taxation of CO₂ emissions and the incentivisation of negative emissions establishing credits for NET. Both approaches have a fundamentally high impact on the economics of all NET [20] and carbon-negative hydrogen production. To provide a basis for initial quantitative assessments on economic prerequisites for HyBECCS, the maximum production costs required for competitive carbon-negative hydrogen are estimated in the following considering external factors. Therefore, the existing estimates of production costs for competing green hydrogen and expected incentives for the active removal of CO₂ from the atmosphere are carried out in the following, using the example of Germany.

The market price for hydrogen is a major external impact factor on the economics of carbon-negative hydrogen. HyBECCS plant operators must be able to produce at costs that are below this market price, which depends, among other things, on the general hydrogen demand and its supply situation. Forecasts predict a sharp increase in hydrogen demand in the coming decades [18,37]. Despite the extensive expansion of electrolysis capacities, there will be a large gap between demand and generation capacity in Germany that is to be closed by imports of green hydrogen according to their national hydrogen strategy, as described in Section 3.1. Therefore, a distinction regarding hydrogen prices in Germany can be made between imported green hydrogen and green hydrogen that is produced in the internal market. Since internal demand is very unlikely to be met by the production of green hydrogen by electrolysis within national borders [18], there will be no direct competition between green hydrogen and carbon-negative hydrogen within Germany. The production costs for carbon-negative hydrogen should therefore be measured against the price of imported green hydrogen. Accomplished studies have estimated the price for hydrogen produced in North Africa and transported to Germany either via pipelines or by ship at 3.10 € per kg in 2050. Despite the lower costs for renewable energy, this is more expensive compared to hydrogen produced in the domestic market (2.45 €). Average

transport costs are estimated at 1.70 per kg H₂ [123]. Other studies come up with supply costs of up to 119 €/MWh with the example of importing green hydrogen from North Africa to Central Europe, which equals 3.97 € per kg H₂ [124]. Further studies predict production costs between 3.85 € and 4.81 € per kg H₂ from solar energy in Morocco [125], amounting to supply costs of 5.55 € and 6.51 € including transport costs, according to [123].

Additionally, credits for CCS should be considered to assess the economics of NET like carbon-negative hydrogen production. The introduction of policy instruments that incentivize NET deployment by generating revenues linked to the amount of CO₂ captured and safely stored is likely to be seen [20]. The expected potential credits per tonne of CO₂ taken from the atmosphere through HyBECCS are estimated in the following. Worldwide, the so-called social costs of carbon (SCC) serve as benchmarks for pricing or tax models. These correspond to the damage caused by an additional tonne of CO₂ to the economy and society and were first defined by Nordhaus [126]. In recent decades, numerous quantitative estimates of SCC have been made. Based on several available models, the National Academies of Sciences, Engineering, and Medicine quantify the SCC for one tonne of CO₂ at an average of 46 € in 2020 and 55 € in 2030 [127]. These figures are based on well-documented empirical assessments of climate damage and can be considered conservative [128]. However, there is uncertainty about the extent of the expected warming, damage, and risks. An expert survey on expected climate damage with 386 respondents from different disciplines placed the average estimate of SCC at 70–90 € per ton CO₂. Other investigations from the German Environment Agency (GEA) consider an SCC of more than 180 € per tonne of CO₂ realistic [129,130]. Governments around the world are basing their statutory CO₂ prices on these calculations. In Germany, for example, since January 2021, the emissions of carbon dioxide in the building and transport sectors have been affected by a fixed price of 25 € per tonne of CO₂, thus covering areas that are not yet affected by the European emissions trading system. This uniform price per tonne of CO₂ emitted is to rise gradually, to 30 € per tonne in January 2022, 35 € in 2023, 45 € in 2024, and 55 € in 2025 [131]. Switzerland, on the other hand, has been charging 96 Swiss francs per tonne of CO₂ since 2018, equivalent to 86.50 €, and has passed legislation that could raise the price to 210 Swiss francs per tonne of CO₂, equivalent to 189.22 € [132].

An estimate of the impact on the production of one kilogram of carbon-negative hydrogen in Germany is based in the following on the assumption that a credit per tonne of CO₂ actively extracted from the atmosphere is made at the level of SCC or existing CO₂ taxes. For this, a worst-case and best-case distinction is made. In the worst-case path considered, we use the agreed price in Germany of 55 € per tonne of CO₂, applicable from 2025 [131], and for the best-case scenario the proposed price of 180 € per tonne of CO₂ of the GEA [130] is assumed. Provided that the use of 1 mol (0.002 kg) of carbon-negative hydrogen is associated with the capture and storage of 1 mol of CO₂ (0.044 kg), the expected credits that could be paid per kg of carbon-negative hydrogen can be estimated. For the worst-case scenario, this results in an incentive of 1.20 €, and for the best-case scenario of 3.93 € per kilogram of carbon-negative hydrogen produced. Also for the supply price of competitive green hydrogen, a distinction is made between a minimum case of 3.10 € and a maximum case of 6.51 € [123,125]. In the figure below, the CCS incentive forecasts, together with the estimated production costs for imported green hydrogen, are shown.

As shown in Figure 4, the incentives for CO₂ capture and storage can have a major impact on the economics of the HyBECCS concept. The sum of the estimated target prize per kg of carbon-negative hydrogen and CCS incentives gives a basic value from which to draw initial conclusions about the maximum production costs of carbon-negative hydrogen to be competitive in the future hydrogen market in Germany. Under the assumptions made, the production costs would have to be below 4.30 € per kg of carbon-negative hydrogen in the worst case and below 10.44 € in the best case to be able to compete with imported green hydrogen. To date, no comparative estimates of the production costs of carbon-negative hydrogen have been available, but this first classification sets a target for cost-effectiveness analyses, which should consider both external and internal impact factors.

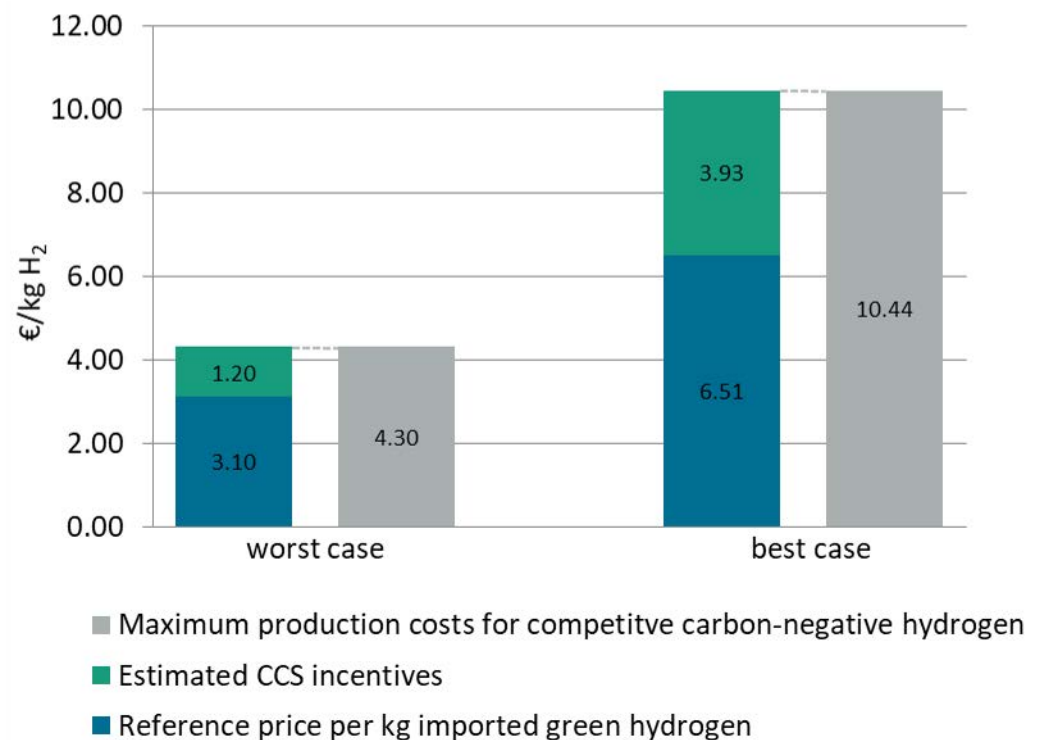


Figure 4. Pricing fundamentals for carbon-negative hydrogen considering the supply costs of competing imported green hydrogen and the expected incentives for CO₂ capture and storage in Germany.

6. Limitations

The HyBECCS concept described in this paper comprises a modular-based system consisting of variable sub-processes. Many of them are already known and tested. However, a complete HyBECCS process combination has never been implemented in a contiguous facility. Their testing in pilot and experimental facilities is still pending and could lead to new findings or process challenges that are not described in this paper. The concept's classifications of ecology (Section 5.1) and economics (Section 5.2) described in this paper are based on simplifications and assumptions made by the authors in an effort to make a conservative estimate. For the ecological assessment, only single-stage dark fermentation, the exclusive use of biogenic residues, a focus on the densely populated country (Germany), and the use of H₂ for the substitution of natural gas were assumed to give a realistic estimate and in order not to create unrealistic expectations. On the other hand, possible emissions connected to farming and the pretreatment of biomass, as well as during carbon storage, which would have an effect on the emission balance, were not taken into account in this paper. In the economic estimation, possible financial benefits through the use of CO₂ as raw material were excluded, as well as possible financial benefits from soil carbon sequestration. Furthermore, carbon-negative hydrogen was compared with imported green hydrogen, although a better carbon footprint of carbon-negative hydrogen is likely to lead to a market advantage over green hydrogen. Nevertheless, process-related peculiarities that are still unknown due to the low technology maturity can lead to negative effects on the economic viability of the HyBECCS approach. The assumptions made need to be reviewed and validated in further research. Additionally, a detailed techno-economic analysis of the process concepts, testing in pilot and experimental plants as well as holistic life-cycle assessments are needed. Then, more precise statements about the potential of carbon-negative hydrogen and the HyBECCS approach will be possible.

7. Summary

In this paper, the HyBECCS (hydrogen bioenergy with carbon capture and storage) concept was introduced as a new approach for NET. This concept combines hydrogen

production from renewable biomass with downstream processes for CO₂ capture and storage or use. The hydrogen produced has a negative carbon footprint and is referred to as carbon-negative hydrogen. HyBECCS processes are described as modular process combination options that can be assigned to four basic steps: the pre-treatment of the substrate biomass, the biohydrogen production process, the separation of hydrogen and CO₂ from the product gas, and their treatment. All steps are interdependent and can be performed by different technology options, some of which are described in this paper. Although HyBECCS has never been technically implemented as a physically connected process combination, there are technology options for each of the four steps that have either already reached market maturity or whose functionality has been proven and tested on a laboratory or prototype scale. The main advantage of HyBECCS over competing NETs can be seen in the efficient capture of CO₂ with high purity produced as point source in a comparatively high concentration during the second step, biohydrogen production. Therefore, technological disadvantages arising either from the contamination of CO₂ with combustion residues or from the energy- and cost-intensive concentration of CO₂ from heterogeneous gas mixtures can be avoided. Further, hydrogen can either be used in the chemical industry, for steel production, or to generate emission-free heat and electricity, in contrast to GHG-emitting hydrocarbon-based energy carriers like biogas or other biofuels produced, for example, from biomass within BECCS approaches. Due to these advantages, the ecological and economic benefits of the HyBECCS concept can be concluded. These benefits were analyzed using the example of Germany. An ecological assessment was made estimating the GHG emission reduction potential through HyBECCS approaches. For the production and use of carbon-negative hydrogen, a theoretical saving potential of 8.49–17.06 MtCO_{2,eq}/a was estimated for the year 2030 in Germany based on existing estimations of biohydrogen production through dark fermentation processes. This results from the potential combination of CO₂ avoidance by substitution of natural gas with hydrogen as an energy source (2.00–4.02 MtCO_{2,eq}/a) and the active extraction of CO₂ from the atmosphere (6.49–13.04 MtCO_{2,eq}/a). In addition, to provide a basis for initial estimates of the economics of the HyBECCS concept, the maximum costs for competitive carbon-negative hydrogen production were estimated for Germany in two scenarios. For this purpose, existing estimations for the production costs of the competing product green hydrogen were added to potential monetary incentives for the extraction of CO₂ from the atmosphere. According to these estimations, the production costs of carbon-negative hydrogen would have to be below 4.30 € per kg of carbon-negative hydrogen in the worst-case and below 10.44 € in the best-case scenario to be able to compete with imported green hydrogen in Germany. In this paper, the definition and differentiation of the HyBECCS concept for sustainable energy solutions as well as initial potential assessments have set the basis for a more in-depth consideration of this novel approach.

8. Outlook

In further investigations, the HyBECCS concept presented in this paper should be analyzed in more detail so as to identify particularly promising process combinations and prepare for experimental research and technological implementation. A holistic techno-economic and ecological understanding of the HyBECCS process's combination possibilities must therefore be gradually developed in order to drive their development and optimization in a targeted manner. To ensure the ecology of the processes, relevant environmental impacts must be identified and analyzed on this basis. A holistic life cycle assessment following DIN EN ISO 14040/44 for the production of carbon-negative hydrogen through HyBECCS should be established. In addition, cross-impact analyses and model calculations on the overall economic implementation in the energy system and industry should be carried out with a focus on intersectoral system developments, e.g., by means of material and energy flow system model analyses. However, to bring carbon-negative hydrogen into widespread use, not only economic and ecological optimization is needed. Social and political framework conditions must also be adapted. A robust

quantification of negative emissions under international supervision is required. Further, social and environmental conflicts, in particular land and water use conflicts, must be prevented. Establishing a certification system for carbon-negative hydrogen, which can ensure that the product has a negative carbon footprint, could help to ensure widespread and long-term acceptance.

From a technical point of view, HyBECCS process combinations can be combined with bioeconomic biorefinery concepts in perspective. An expanded range of products and substrates could lead to higher value creation and improve economic efficiency. The main product gas fractions H_2 and CO_2 could therefore be used in a combinatorial way, for example by using H_2 as a reducing agent for CO_2 , as outlined in Figure 5. Eco-efficient carbon cycles with high added value can thus be created based on the presented HyBECCS approach.

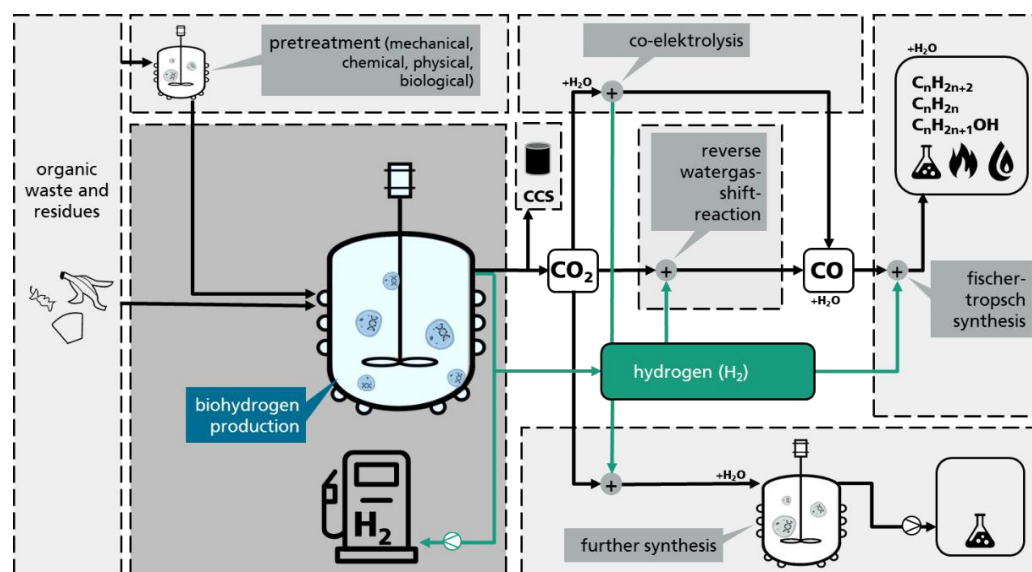


Figure 5. Exemplary scheme for the expansion of the product range to create a biorefinery concept.

However, the most important basis for both HyBECCS and bioeconomy approaches is a deeper techno-economic understanding as well as a comprehensive life-cycle assessment of the basic process step of biohydrogen production from biomass.

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