# Renewed perspective on the role of biogas as local and or regional source of bio-hydrogen and bio-carbon dioxide within the future renewable hydrogen energy system

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# Renewed perspective on the role of biogas as local and or regional source of bio-hydrogen and bio-carbon dioxide within the future renewable hydrogen energy system

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# Abstract

A radical change of the energy system is required in light of the dramatic effects of human-induced climate change. In this perspective, a future renewable hydrogen energy system was proposed, where biogenic resources are ascribed relevant potential as source of biogenic hydrogen and biogenic carbon dioxide. In this light, the research brings forward the concept of third-generation upgrading as the highest valorisation potential of biogas. This alters the perspective on biogas as source of renewable electricity, heat or fuel to biogas as platform molecule able to couple the renewable hydrogen and bio-economy domain. The research shows that concept of third-generation upgrading relies on a technologically feasible, environmentally benign and economically sound production process. In this respect, the adoption of the concept of third-generation upgrading is supported via the valuation of the bio-carbon dioxide as scarce renewable carbon-containing molecule and valuable carbon sink. Therefore, the concept of third-generation upgrading should be supported via regulatory commitment, market creation, policy mechanisms and infrastructural changes. In this way, biogas can act as an invaluable source of bio-hydrogen and bio-carbon dioxide, spark system integration and stimulate energy security. Ultimately, this research proposes to change the way biogas is seen.

*Keywords:* Biogas, Hydrogen, Carbon, Carbon Dioxide, Renewable Energy System 2022 MSC: TBD, TBD

# 1. Introduction

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There is an immediate urge for climate action in order to re-<sup>25</sup> spond to the dramatic effects of human-induced climate change. The time frame for interference is shortening, while the unprecedented changes to the climate are accelerating. These changes to the climate are already affecting extremes in weather and climate, are expected to become larger and many changes <sup>30</sup> are irreversible [1] [2]. Therefore, decisive action is required to establish deep reductions in carbon dioxide and other greenhouse gases as the costs of inaction are immense and the risks

and stakes are high: "It is a code red for humanity" [3] [4]. This calls for a far-reaching transformation of the economic <sup>35</sup> system including the power, industrial, transportation and agricultural sectors [2]. The European Green Deal, in line with the

Paris Agreement, aims for a climate-neutral energy system by 2050. This will fundamentally transform the economy and society towards a fair, green and prosperous future [5]. This radi-40 cal change will require a mentality shift towards embracing the need for radical transformations as incremental changes alone will not be sufficient [6].

The urgent need to transform the energy system sparked renewed momentum for the utilisation of renewable hydrogen as  $_{45}$ 

carbon neutral and zero pollution energy vector [7] [8]. Renewable hydrogen will be, next to large scale electrification and circular usage of materials, vital to achieve a climate neutral energy system by 2050 [9]. Renewable hydrogen will allow for the cost-effective transport and storage of cheap renewable electricity over time and space [10]. Moreover, hydrogen will allow for the integration of the power, industrial, transportation and industrial sectors. In this respect, hydrogen can act as energy vector to decarbonise hard-to-abate sectors, balance the power sector or could be used to decarbonise processes, products and materials [11]. In this way, renewable hydrogen will be central in a climate-neutral energy system [12].

The proposed future renewable hydrogen energy system shows important similarities to the present natural gas system. Hydrogen will fulfill the role of zero pollution energy commodity that facilitates connection over time and space dimensions [13]. In this way, hydrogen will replace fossil fuel utilisation and contribute to an affordable, accessible, secure, reliable and fair transition [14]. Moreover, the proposed renewable hydrogen system will support the reduction of greenhouse gas emissions, the diversification and security of the energy supply, the integration of renewables and will spark economic growth and technology development. The proposed future renewable hydrogen energy system can be observed in figure 1 [13].

In the proposed renewable hydrogen energy system, biogenic resources constitute a vital role as local and or regional source of biogenic hydrogen and biogenic carbon dioxide. In this role, the biogenic hydrogen can be fed into the hydrogen infrastruc-

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Figure 1: Schematic lay-out of the proposed future renewable hydrogen system

ture to be used as energy carrier or feedstock while the biogenic carbon dioxide can be utilised, for example, as building block in the industrial sector or as product in the horticulture sector. The intermediate synthetic gas could also directly be used as circular chemical feedstock [13]. In this perspective, biogenic resources are seen as a vital source of climate neutral carbon

dioxide in the renewable hydrogen energy system.

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Biogas, as wet biogenic resource, has been ascribed as an attractive source of bio-hydrogen from a technological [15], eco-

- <sup>60</sup> logical [16] and economical perspective [17]. Nevertheless, the<sub>115</sub> focus on biogas has been primarily on the direct energetic utilisation for power and or heat production or on the indirect energetic utilisation as green gas in the industrial, transportation and or build environment sector [18]. However, next to the con-
- tribution to the renewable energy domain, biogas has also been<sub>120</sub> ascribed potential within the bio-economy domain as input for various bio-based products to replace fossil counterparts. Based on the cascading principle, biogas is ascribed a dual and time-dependent role in both transitions [19]. In this respect, biogas
- <sup>70</sup> is seen as platform molecule of both green gas and bio-carbon<sub>125</sub> dioxide. In the concept of second-generation upgrading of bio-gas, the separated bio-carbon dioxide is not disregarded but assigned a potential for the production of hydrocarbon-based high energy-density fuels that enables the closure of carbon cycles.
- <sup>75</sup> This offers a flexible storage option important within an inter-130 connected energy infrastructure [20]. However, this perspective lacks the potential of biogas to operate as source of zero pollution bio-hydrogen and the complete, circular, utilisation of the inherent bio-carbon dioxide.
- Thus, the current vision on biogas has been limited to the<sup>135</sup> direct or indirect energetic utilisation of biogas. This ignores the potential of biogas as valuable source of bio-hydrogen and bio-carbon dioxide in a fossil free energy system. Therefore, in light of the renewable hydrogen energy system the concept of
- third-generation upgrading is proposed as highest valorisation potential of biogas. This changes the way biogas is seen. 140

#### 2. Research Scope

The concept of third-generation upgrading renews the vision on the utilisation of biogas within the proposed renewable hydrogen energy system. In this respect, the concept of thirdgeneration upgrading can be seen as an extension of the historical development of the perspective on biogas utilisation. Biogas was traditionally seen as a renewable electricity, heat or fuel source due to the short-cycled nature of the emitted carbon dioxide. The concept of second-generation upgrading opened the perspective on the potential circular or climate negative utilisation of part of the captured atmospheric carbon dioxide. Now, the concept of third-generation upgrading envisions biogas as source of climate neutral bio-hydrogen and fully exploits the carbon content for circular or climate negative bio-carbon dioxide utilisation as can be seen in figure 2. In this way, biogas not only acts as an important carbon sink but also as a valuable source of bio-carbon within a fossil free energy system.

Therefore, biogas should be seen as a platform molecule able to connect the renewable hydrogen and bio-economy domain. This is based on four perspectives: the bio-hydrogen, bio-carbon, system integration and energy security perspective.

#### 2.1. Bio-hydrogen perspective

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In the renewable hydrogen energy system, the direct utilisation of biogas and green gas is devalued against the utilisation of renewable hydrogen. This is a result of reshaped boundary conditions, like infrastructural changes, policy support mechanisms and technological development. In this way, the upgrading of biogas to bio-hydrogen results in direct economic benefits. Therefore, the concept of third-generation upgrading could better the biogas plant economics [21] [22]. Additionally, the conversion of biogas to bio-hydrogen facilitates the repurposing of the current natural gas infrastructure. This stimulates a fast transition, avoids stranded assets and reduces a potential natural gas infrastructure lock-in [9]. Moreover, hydrogen could be valued as a more versatile renewable molecule with usage in high-value industrial applications. Next to that, the utilisation of hydrogen could see efficiency gains, for example, in fuel cell technology as compared to combustion engines [23]. Lastly, the conversion of biogas unlocks additional renewable hydrogen production capacity, where biogas or green gas can act as a replacement of natural gas in the production of hydrogen [24].

As a result, the concept of third-generation upgrading assigns high economic value to bio-hydrogen production in the short to medium term. This results from insufficient renewable hydrogen production capacity and is supported by infrastructural and regulatory boundary conditions. However, in light of the continued increase in the availability of renewable hydrogen, especially from locations with cheap renewable electricity resources, the relative value of bio-hydrogen is expected to decrease. The commodity value of renewable hydrogen supports the bio-carbon perspective over the longer term.

#### 2.2. Bio-carbon perspective

The concept of third-generation upgrading assigns value to the inherent presence of bio-carbon in biogas. This is a result



Figure 2: Visualisation of the concept of third-generation upgrading based on CO2 utilisation and emission as opposed to previous generations of biogas upgrading

of the need for negative emissions and scarce renewable carbon<sup>170</sup> containing molecules. In the future renewable energy system, sources of carbon are limited as a result of the absence of fossil resources. Here, carbon dioxide is no longer a waste product

- <sup>145</sup> of the energy system [25]. Instead, the renewable energy system is based on electrons, where renewable hydrogen is utilised<sup>175</sup> to transport cheap electrons over the time and space [13]. As a result, scarce renewable carbon containing molecules should primarily be utilised as feedstock and only be used for energetic
- ends if no other option exists. Bio-carbon will become vital for the production of products and materials within the future renewable hydrogen energy system [25]. Therefore, the complete<sub>180</sub> utilisation of the inherent bio-carbon in biogas supports the cascading principle of the concept of third-generation upgrading.
- <sup>155</sup> The concept of third-generation upgrading also assigns value to negative carbon dioxide emissions. The negative carbon dioxide emissions result from the storage and or, circular, utili-<sub>185</sub> sation of bio-carbon dioxide [26]. In this way, biogas is not seen as a carbon neutral energetic resource but as a necessary carbon
- sink able to close the carbon cycle. This is based on the condition that the carbon is captured within products or materials, not released within its lifetime and or is used circularly [27]. This<sub>190</sub> is in contrast to, for example, the energetic utilisation of the captured bio-carbon dioxide in hydrocarbon-based high energy density fuels [20]. Therefore, the concept of third-generation
- upgrading allows biogas to operate as a cost-effective carbon capture technology [28].

Thus, the concept of third-generation upgrading values assigns value to bio-carbon dioxide in terms of the physical delivery of bio-carbon dioxide and the closure of captured atmospheric carbon dioxide. In this respect, the value of biocarbon dioxide is presumed to continue to increase in reaction to the shift from a molecular-based energy system towards an electron-dominated energy system. On top of that, the value of negative carbon emissions are assumed to remain relevant due to the absolute atmospheric carbon balance and lack of feasible alternatives. In this perspective, the relative value of biocarbon dioxide supports the valorisation potential of the concept of third-generation upgrading.

#### 2.3. System integration perspective

In the proposed renewable hydrogen energy system, biogenic resources are discussed to be a relevant local and or regional source of biogenic hydrogen and biogenic carbon dioxide [13]. This is supported by potential delivery cost reductions as result of lower infrastructural requirements. This reduces the relative cost position of bio-hydrogen. In this respect, the system integration potential is dependent on factors like biogas potential, production capacity, industrial demand centers and the current infrastructural network [29]. Next to the relative cost position, the system integration potential could impact the product mix of the concept of third-generation upgrading. For example, production locations in close proximity to industrial demand centers could benefit from the direct utilisation of the produced syngas. In this way, the concept of third-generation upgrading supports local and or regional system optimisation.

For that reason, the concept of third-generation upgrading focuses, besides repurposing of the natural gas infrastructure,

on system integration optimalisation to reduce the infrastructural requirements. This could be supported through the use of

local and or regional production and demand hubs [30]. In this way, the agricultural sector could be coupled with the industrial, power and or transportation sector. This reduces the burden on infrastructural development, supports an affordable transition and stimulates the adoption of the concept of third-generation
 upgrading of biogas.

#### 2.4. Energy security perspective

The concept of third-generation upgrading focuses on short supply chains to boost energy security and enable a secure, reliable and accessible energy transition. This becomes in-<sup>250</sup> creasingly relevant in light of the vulnerabilities associated with globalised and concentrated energy supply chains [31]. Moreover, the focus on local optimisation and short supply chains could also stimulate energy security within small scale and or standalone systems. In this respect, small scale systems could<sup>255</sup>

<sup>215</sup> become energy independent through the use of bio-hydrogen for heat, fuel and or feedstock requirements. This is supported via the utilisation of a widely available problematic waste input stream for biogas production. This could also be used to enable social development of rural communities [22].

Therefore, the concept of third-generation upgrading supports local and or regional energy security and could spark social development. This fits within the wider renewable hydrogen energy system and allows for a secure, reliable and accessible transition.

# 225 3. Results

The concept of third-generation upgrading changes the way<sub>270</sub> biogas is seen. The conversion of biogas to bio-hydrogen and bio-carbon dioxide is ascribed the highest valorisation potential due to production of zero pollution bio-hydrogen and the com-<sup>230</sup> plete, circular, utilisation of the inherent bio-carbon dioxide. Thereby, the concept of third-generation upgrading is supported<sub>275</sub> by a technologically feasible, environmentally benign and economically supportive production process. The concept of third-generation upgrading is placed in the context of the different generations of biogas upgrading.

# 3.1. Technological

Table 1 shows the production steps required for the different generations of biogas upgrading. Moreover, table 2 shows the key process related parameters for the different generations of biogas upgrading.

#### 3.1.1. Conversion

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In table 1 it can be observed that all generations of biogas upgrading require the anaerobic digestion of manure to produce biogas. Anaerobic digestion (AD) is a biochemical process that converts organic matter under depleted oxygen conditions to a gaseous mixture consisting mainly of methane ( $CH_4$ ) and carbon dioxide ( $CO_2$ ) [24]. The resulting biogas consists, depending on the organic waste composition and reactor conditions, of

Production	Zero	First	Second	Third
Anaerobic	Yes	Yes	Yes	Yes
digestion				
Biogas	No	Yes	Yes	Yes/No
upgrading				
Reforming	No	No	No	Yes
technology				

Table 1: Technological production steps required for different generations of biogas upgrading

 $CH_4$  (50-70 v%),  $CO_2$  (30-50 v%) and trace species like nitrogen, hydrogen sulphide, ammonia, oxygen and hydrogen. Biogas has, based on the methane content, a lower calorific value of 20-25 MJ/Nm<sup>3</sup> biogas as compared to around 36 MJ/Nm<sup>3</sup> natural gas [32]. Moreover, next to biogas, a digestate by-product is produced, in the AD process, which is rich in nutrients and could be used as bio-fertiliser, animal bedding and or other horticulture products [22].

Moreover, table 1 indicates that a biogas upgrading production step is required to convert biogas to green gas. The biogas upgrading production step is preceded by a contaminants removal product step to remove contaminants, like hydrogen sulfide, water, ammonia and siloxanes, that have a detrimental effect on the downstream production process, including catalyst poisoning, conversion efficiencies, depositions and corrosion [33]. A wide range of contaminants removal technologies are possible, including absorption, adsorption, membrane separation and biological systems [34]. The exact, combination of, removal technology is in general site dependent due to factors like contaminants present, flow rates and process adaptability. However, in basis activated carbon is mainly used to remove hydrogen sulfide, and siloxanes, while water, and ammonia, is adsorbed on silica gel [35] [36]. Hereafter, the biogas upgrading production step is deployed to separate  $CO_2$  and  $CH_4$  in order to arrive at a natural gas quality green gas. The separation occurs based on the different chemical and or physical behaviours of  $CH_4$  and  $CO_2$  via either membrane technology, pressure swing adsorption, cryogenic technology or scrubbing technology [35]. In case of membrane technology, a multi-stage membrane process is used to separate the  $CH_4$  and  $CO_2$  based on the difference in permeability of both constituents driven by a concentration differences. Here, effective separation occurs as  $CO_2$  permeates through the membrane while the  $CH_4$  retains on the inlet side, resulting in high methane purity and recovery [35]. At the moment, membrane separation is the dominant upgrading technology due to the simplicity of operations, low costs, low energy usage and environmental friendliness [37].

Finally, table 1 shows that the production of bio-hydrogen and bio-carbon dioxide require the reforming of green gas or biogas. In this respect, green gas serves as chemical identical replacement of natural gas in the traditional reforming process [38]. In the commercial steam methane reforming (SMR) process, hydrogen ( $H_2$ ) is obtained via subsequent oxidation of methane with steam in the SMR reaction to obtain a syngas, the oxidation of carbon monoxide (CO) with steam in the water gas shift reaction (WGSR) to increase the hydrogen content,

and the separation of  $H_2$  and  $CO_2$  via pressure swing adsorption (PSA) to adhere to the hydrogen purity requirements [39] [40]. The SMR process is the dominant industrial hydrogen production technology due to the high hydrogen yield, the process reliability and low cost perspective [41] However, also alternative process are possible that focus on the utilisation of alternative oxidation agents [17] [37]. In this respect, alternative reforming reactions are partial oxidation reforming (POX) and dry reforming (DR) that utilise oxygen and carbon dioxide as oxidant respectively. Moreover, autothermal reforming (ATR) combines the exothermic POX and the endothermic SMR reactions [42]. ATR has been ascribed additional benefits in relation to the lower process energy requirement, controllable hydrogen over carbon ratio, ease of operations, high efficiency and high<sup>310</sup> hydrogen yield, despite a more complex reactor design, oxygen input requirement and higher operating pressure and temperature [39]. The ultimate reaction mechanisms in the ATR, where POX and SMR occur consecutively in the same reactor, can be 315 summarised as follows: [39]:

$$CH_4 + \frac{1}{2}O_2 \longrightarrow CO + H_2O$$
 (-519 kJ/mol) (1)

$$CH_4 + H_2O \longrightarrow CO + 3H_2 \qquad (+206 \ kJ/mol) \qquad (2)^{326}$$

The subsequent WGSR can be summarised as follows [39]:

$$CO + H_2O \rightleftharpoons CO_2 + H_2 \qquad (-41 \ kJ/mol) \qquad (3)$$

For the final  $H_2$  purification step several hydrogen separation<sup>325</sup> methods exist that can be characterised as either physical methods, including PSA, cryogenic distillation, and membrane separation and chemical methods like metal hydride separation [40] [43] [44]. Nevertheless, PSA, a semi-continuous process that relies on the static capacity difference of hydrogen in relation<sup>330</sup> to other majority gas molecules, is the industrial dominant separation technology due to the high  $H_2$  purity requirement, low operational costs and long service life [40].

Next to the traditional reforming layout, several alternatives surface that focus on the intensification of the conversion process through elimination of the biogas upgrading step and or hydrogen purification step [45]. In this respect, biogas reforming could be characterised as DR due to the  $CO_2$  levels naturally present in the biogas and could be stated as [42] [46]:

$$CH_4 + CO_2 \longrightarrow 2CO + 2H_2 \qquad (+247.2 \ kJ/mol) \qquad (4)^{340}$$

- In typical DR reactions, a more useful syngas could be obtained due to the lower concentration levels of *H*<sub>2</sub> and higher levels of *CO* present [33]. However, due the unfavourable carbon deposition and low hydrogen yield is DR not applied [47] [48]. In re-<sub>345</sub> sponse, a combination of oxidants is utilised to lower the carbon deposition and enhance the hydrogen yield. These include biogas dry reforming [49] [50], as combination of oxygen and carbon dioxide, and biogas autothermal reforming [52]<sub>350</sub> [53] [54], as combination of steam, oxygen and carbon dioxide.
- <sup>305</sup> This showed the potential to prevent carbon decomposition,

Variable	Zero	First	Second	Third
Energy vector	Biogas	Green gas	Green gas	Hydrogen
Volume $(Nm^3)$	0.84	0.6	0.6	1.1
Energy LHV (MJ)	18.1	21.6	21.6	12.1
Carbon yield (kg)	0	0	0.44	1.36

Table 2: Key process related variables for different generation of biogas upgrading per  $Nm^3$  biogas input

regulate the hydrogen over carbon ratio, increase the hydrogen yield and obtain almost complete methane and carbon monoxide conversion. Nevertheless, a higher  $CO_2$  content and a lower  $H_2$  yield could be obtained due to unfavourable WGSR equilibrium conditions [46]. Other process intensification methods aim to simplify the reaction process through the integration of the hydrogen production and separation step. These process intensification methods offer the potential to lower the process complexity at high hydrogen yield. The dominant process methods are steam-iron reforming [55] [56], membrane reforming [57] [58] [59] [60], sorption enhanced reforming [61] [62] and methane cracking [63].

Overall, ATR related conversion methods are presumed to have the highest potential for the conversion of biogas within the proposed renewable hydrogen energy system. This is related to the high level of system flexibility, production scalability and process output. This is further supported by the high level of technological feasibility, high carbon capture rate potential and the low cost perspective. Biogas ATR might prove to be most useful while membrane reforming might be more specific for instances with high hydrogen output requirements. In contrast, SMR and methane cracking might be less relevant for the concept of third-generation upgrading due to the high production scale and low system flexibility. Lastly, the process intensification options of sorption enhanced reforming and steam-iron reforming could prove to become important over the medium to long term as a result of the low technological readiness level. In this respect, the research focuses on the traditional reforming process and the innovative biogas ATR process.

Therefore, it can be seen that the concept of third-generation upgrading requires an additional, commercially available, green gas reforming step. Nonetheless, process intensification via biogas ATR has the potential to exclude the biogas upgrading step and lower the system complexity and cost.

## 3.1.2. Process

In table 2 it can be seen that the energy content decreases over biogas upgrading generations while the carbon yield per  $Nm^3$ biogas increases. This can be explained by non-energetic benefits over upgrading generations via, for example, increased energy flexibility, technological applications and political support [26]. Moreover, the higher carbon yield supports an adequate valuation of the inherent bio-carbon dioxide via, for example, negative  $CO_2$  emissions and or bio-carbon dioxide applications.

The conventional bio-hydrogen production process could be described as biogas production (0nd generation upgrading), biogas upgrading (1st and 2nd generation upgrading) and green gas reforming (3rd generation upgrading). First, the biogas pro-

Production	Zero	First	Second	Third	
Waste man-	Yes	Yes	Yes	Yes	
agement					
Methane	Yes	Yes	Yes	Yes	395
emissions					395
Water, soil,	Yes	Yes	Yes	Yes	
air pollution					
Bio-digestate	Yes	Yes	Yes	Yes	
production					
Energy sys-	Limited	Yes	Yes	Yes	400
tem flexibility					
Carbon diox-	Ne	No	Limited	Yes	
ide capture					

Table 3: Overview of the environmental benefits for different generations of biogas upgrading  $^{\rm 405}$ 

duction yield is around 25  $Nm^3$  biogas/tonne cow manure in case of mono-digestion [64] [65]. However, based on a presumed internal heat demand of the digester of 16%, approx-<sup>410</sup> imately 0.84  $Nm^3$  biogas could be obtained per  $Nm^3$  biogas [64]. The subsequent upgrading of biogas, of approximately  $60\% CH_4$  and  $40\% CO_2$ , to green gas, of approximately 80% $CH_4$  and  $20\% CO_2$ , lowers the output product volume by an approximate factor of 70%. The resulting loss in production<sup>415</sup> volume is recovered in the form of a bio-carbon dioxide stream of around 0.44 kg  $CO_2/Nm^3$  biogas. This is based on a  $CH_4$ loss of *conf.*, a  $CO_2$  loss of *conf.* and the assumption that exter-

- nal renewable electricity is provided [26] [35]. Hereafter, based on an approximate 30% internal heat demand, 90%  $CH_4$  con-<sup>420</sup> version, 95% CO conversion and 85%  $H_2$  recovery rate, a highpurity bio-hydrogen stream of 1.1  $Nm^3/Nm^3$  biogas is obtained [17] [40] [50] [65]. Moreover, based on a process capture rate of 90% [66], an additional 0.93 kg  $CO_2/Nm^3$  biogas stream can
- <sup>370</sup> be recovered in the reforming process. Here, around 0.29 kg<sup>425</sup>  $CO_2/Nm^3$  biogas could be obtained from the fuel gas and 0.64 kg  $CO_2/Nm^3$  biogas from the process gas respectively. However, in the case of the biogas ATR layout a 5-10% decrease in hydrogen yield to 0.085 kg  $H_2$  or 1.01  $Nm^3 H_2/Nm^3$  biogas could be observed as a result of the lower stoichiometric hydro-<sup>430</sup>

gen output. Nonetheless, an increase of over 10% in bio-carbon dioxide to 1.54 kg carbon dioxide/Nm<sup>3</sup> biogas could be seen. This ultimately yields an approximate decrease in energy content of 45% but a factor 3 increase in carbon yield in case of third-generation upgrading as opposed to the concept of second<sup>435</sup> generation upgrading.

Therefore, the concept of third-generation upgrading shows longer energetic value as compared to previous generations of biogas upgrading. Nevertheless, the concept of third-generation upgrading shows a significant increase in bio-carbon dioxide<sup>440</sup>

output that reinforces the cascading principle.

#### 3.2. Environmental

Table 3 shows the environmental benefits for different gener-445 ations of biogas upgrading.

In table 3 it can be observed that most environmental benefits are related to the production of biogas. The production of biogas is considered an established sustainable process for the simultaneous generation of renewable energy and treatment of, problematic, organic waste [24] [26]. It is identified that AD is the preferred waste to energy route [22]. Thereby, monodigestion of manure offers additional benefits related to lower methane leakage, other water, soil and air pollutants and odour pollution [21] [67]. Moreover, the by-product digestate could lower nutrient runoff, avoid methane emissions, conserve soil quality and ultimately adhere to the circularity principle [68]. In this respect, the sequestration of the process digestate could result in net negative process life cycle  $CO_2$  emissions [44]. On top of that, AD could offer a solution to the nitrogen crisis [69]. Lastly, in case of AD problems associated with land use change, land availability and food competition are circumvented and overall the usage of manure shows the strongest positive climate impact [70] [71].

Moreover, in table 3 it can be seen that positive effects of increased energy flexibility are associated with the upgrading of biogas. This relates to less renewable electricity curtailment, as a result of the better storage potential of green gas [26]. Additionally, in table 3 it can be observed that second-generation upgrading and, especially, third-generation upgrading offers direct benefits related to the potential to capture, and utilise, the inherent bio-carbon dioxide. In this respect, biogas could be considered a carbon capture technology of both atmospheric  $CO_2$  and  $CO_2$ -equivalent emissions that would otherwise have been released to the air [20]. On the other hand, the utilisation of green gas is characterised by similar  $CO_2$  emissions as fossil counterparts, despite considered carbon neutral [35] [72]. This reduces the potential to obtain negative  $CO_2$ -equivalent emissions in case of second-generation upgrading. Moreover, the exact utilisation of the captured bio-carbon dioxide is important. In this respect, the utilised  $CO_2$  is considered small-cycled in case of food applications as the  $CO_2$  is presumed to be released within weeks. This is extended to months in case of application in chemicals or fuels. In case of  $CO_2$  usage in plastics and or minerals the  $CO_2$  is presumed to be captured over (tens of) years [27]. This in turn puts emphasis on the circular utilisation of bio-carbon dioxide with preferred use cases as bio-feedstock to obtain net negative process emissions. Therefore, through the capture and subsequent storage or circular utilisation of process and product  $CO_2$ , the concept of third-generation upgrading shows net negative process emissions [44]. In this way, the concept of third-generation upgrading could contribute to the carbon economy by closing the carbon loop.

The concept of third-generation upgrading is ascribed environmental benefits as source of zero pollution renewable hydrogen and as, potential, negative carbon source. In the short term, the role of biogas as carbon sink is seen vital to not overshoot the global carbon balance [28]. In the longer term, the potential circular utilisation of bio-carbon dioxide is seen vital within a fossil free and electron dominated energy system [25]. As a result, a carbon mass analysis is deployed to quantify the environmental benefits of the concept of third-generation upgrading in relation to previous generations of biogas upgrading.

Figure 3 portrays the carbon balance over the different steps in the traditional bio-hydrogen production process, based on similar process assumptions as mentioned in section 3.1.2. Fig-

<sup>450</sup> ure 3 excludes other relevant environmental benefits and drawbacks associated with biogas production. It also excludes potential carbon sequestration via bio-fertiliser utilisation. In this respect, around 40-60% of the input organic matter ends up in the bio-fertiliser out of which around 30% ends up in the

soil [44]. On the other hand, negative effects, like transportation and storage related *CO*<sub>2</sub>-equivalent emissions, are also excluded. In this respect, the integration with renewables could lower the downstream impact. On top of that, figure 3 presumes renewable energy integration throughout the production process, which includes electrical energy consumption in case of membrane separation [73] and the deployment of renewable electricity and heat in the reforming, bio-hydrogen separation and carbon capture steps [65] [74].

In Figure 3 it can be observed that an initial 46.7 kg *CO*<sub>2</sub> equivalent is locked in 25 *Nm*<sup>3</sup> biogas produced or 1.9 kg *CO*<sub>2</sub>/*Nm*<sup>3</sup> biogas. This is based on the theoretical complete combustion of *CH*<sub>4</sub> to obtain *CO*<sub>2</sub>, or around 2.7 kg *CO*<sub>2</sub>/kg<sup>505</sup> *CH*<sub>4</sub>. Ultimately, 10.9 kg *CO*<sub>2</sub> could be recovered in the concept of second generation upgrading. In case of thirdgeneration upgrading an additional 23.2 kg *CO*<sub>2</sub> could be obtained to increase the total carbon capture potential to 34.1 kg or 1.4 kg *CO*<sub>2</sub>/*Nm*<sup>3</sup> biogas. A total of 7.4 kg *CO*<sub>2</sub>-equivalent is<sup>510</sup> lost due to internal heat demand in the biogas production step, 0.2 kg *CO*<sub>2</sub> due to losses in the biogas upgrading step and 5.0

 $kg CO_2$  as a result of the imperfect capture rate in the reforming step. In case of the biogas ATR process the approximate theoretical carbon capture yield would be 1.53 kg  $CO_2/Nm^{3515}$ biogas or a total of 38.4 kg  $CO_2/t$  manure. As a result, the biogas ATR reaction shows an approximate 15% increase in biocarbon dioxide capture potential as compared to the reference installations, despite the potential lower hydrogen yield.

Therefore, the concept of third-generation upgrading shows<sup>520</sup> additional environmental benefits in relation to the carbon capture potential of biogas both in absolute terms and in reference to the concept of second-generation upgrading. In this respect,

to the concept of second-generation upgrading. In this respect, the concept of third-generation upgrading has the potential to increase the bio-carbon dioxide capture potential with a factor<sub>525</sub>
3. In this way, the concept of third-generation upgrading has the potential to unlock the value of bio-carbon dioxide and contribute to a circular carbon economy.

#### 3.3. Economical

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Table 4 shows the relevant economic parameters for the different generations of biogas upgrading. Table 5 shows the levelised cost figures for the different generations of biogas upgrading. Finally, table 6 indicates the relevant process, production and costs assumptions underlying the calculations.

In table 4 it can be observed that the production costs of the different generations of biogas upgrading is mainly determined by the biogas input price. In the case of green gas production, the biogas input price accounts for over *conf.* of the total unit cost price, while for the traditional third-generation upgrading<sup>540</sup> layout the biogas input price accounts for over *conf.* of the total unit cost price. This highlights the importance of feed-stock costs in case of reforming technologies [66] and implies

Variable		Zero	First	Second	Third
Output pr	oduct	Biogas	Green gas	Green gas	Hydrogen
Output un	it	$Nm^3$	$Nm^3$	$Nm^3$	kg
Input (EUR)	Costs	0	0.36	0.36	2.34
CAPEX alised (EU	annu- JR)	N/A	conf.	conf.	1.22
OPEX an (EUR)	nualised	N/A	conf.	conf.	0.54
Total Un (EUR)	it Cost	0.25	conf.	conf.	4.11
Unit cost biogas)	$(\in/Nm^3)$	0.25	conf.	conf.	0.44

Table 4: Key process related variables per output unit for different generation of biogas upgrading per  $Nm^3$  biogas input based on 5.5 MW biogas installation

the lower economic performance of bio-hydrogen production in relation to traditional lower cost natural gas reforming [37]. The higher cost position as compared to traditional natural gas reforming can also be explained by lower economies of scale in case of bio-hydrogen production. This relates to the strong CAPEX contribution of more than double the OPEX contribution [66] [75]. In contrast, the 5.5 MW biogas production reference installations shows important economies of scale in the biogas upgrading plant due to the relatively large-scale capacity [64]. Overall, the production cost in case of second-generation upgrading is *conf.*  $\in$ /Nm<sup>3</sup> green gas and for third-generation upgrading is 4.11  $\in$ /kg  $H_2$ , or 3.61  $\in$ /kg hydrogen excluding carbon capture and storage technology. However, in case of biogas ATR, the unit cost of bio-hydrogen production could be reduced to 3.81  $\in$ /kg  $H_2$  and 3.31  $\in$ /kg  $H_2$  respectively as a result of process integration and similar cost structure, despite the lower hydrogen yield and higher feedstock contribution.

Moreover, in table 4 it can be seen that the unit cost per biogas input increases over the generations of biogas upgrading. In this respect, the biogas upgrading adds approximately conf. to the unit cost price, while the green gas reforming adds around an additional conf. As a result, the concept of third-generation upgrading adds around *conf.*  $\in$ /kg  $H_2$  to the second-generation upgrading to produce bio-hydrogen and bio-carbon dioxide. The bio-hydrogen yield of the concept of third-generation upgrading is around 1.2-1.3 Nm<sup>3</sup> H<sub>2</sub>/Nm<sup>3</sup> biogas and 1.5-1.6 kg  $CO_2/Nm^3$  biogas in contrast to a green gas yield of 0.7  $Nm^3$ green gas/ $Nm^3$  biogas and 0.5 kg  $CO_2/Nm^3$  biogas. As a result, an approximately double normal volume amount of biohydrogen per normal volume amount of green gas and three times the amount of bio-carbon dioxide should recover the conf.  $\in$ /kg  $H_2$  increase in production costs for the concept of thirdgeneration upgrading. However, the energy content of hydrogen per normal volume as compared to natural gas equivalent is around 25-30% [76], which results in an energetic value decrease of around 45-55% of the bio-hydrogen output in comparison to the green gas output. On the other hand, higher downstream application efficiencies, of 20-50% [23], would effectively reduce the energy disparity to 25-40% in disfavour of bio-



Figure 3: Carbon mass (equivalent) balance analysis over the different traditional bio-hydrogen production steps in  $Nm^3$  (Green arrows indicate carbon capture potential and red arrows indicate potential carbon losses. BG is biogas and GG is green gas.)

Production	Zero	First	Second	Third
LCOE (€/MWh)	50	conf.	conf.	130-145
LCOH ( $\in/kg H_2$ )				4.4-4.9

Table 5: Dominant levelised cost figures for the different generations of biogas

upgrading

The LCOH is derived from equation 5 [17] [79].

$$LCOH = \frac{(TPC * CCF) + C_{O\&M_{fix}} + C_{O\&M_{var}}}{kg_{H_2}}$$
(5)

The TPC can be determined by equation 6 [17].

$$TPC = \left[ \left( \Sigma C_{i,2022} * (1 + \mathcal{H}_{TIC}) \right) * (1 + \mathcal{H}_{IC}) \right] * (1 + \mathcal{H}_{C\&OC})$$
(6)

hydrogen. Therefore, the additional captured bio-carbon dioxide in the concept of third-generation upgrading should be able to recoup an additional *conf*.  $\in$ /kg hydrogen production costs and an energetic value loss of around *conf*.  $\in$ /kg. Therefore,<sub>570</sub> based on a bio-carbon dioxide yield of around 15 kg *CO*<sub>2</sub>/kg *H*<sub>2</sub> as opposed to 5 kg *CO*<sub>2</sub> in the concept of second-generation upgrading, a theoretical bio-carbon dioxide value of 200-275 EUR/t *CO*<sub>2</sub> would support a positive cost position of the concept of third-generation upgrading.

Table 5 indicates that the levelised cost of hydrogen (LCOH) of the biogas ATR and traditional steam green gas reforming is  $4.4-4.9 \in /kg H_2$  or equal to a levelised cost of energy (LCOE) of 130-135  $\in /MWh$ . The higher LCOH as compared to the unit cost could be explained by the different assumptions with respect to the total plant costs (TPC). Additionally, in table 5 it can be observed that the LCOE for the third-generation upgrading *conf.* as compared to the concept of second-generation

- <sup>560</sup> grading *conf.* as compared to the concept of second-generation upgrading. The relative lower difference in energetic value between the second and third-generation upgrading relates to<sup>585</sup> assumptions with respect to the energetic content of the output products and the TPC. Nevertheless, the LCOE results
   <sup>565</sup> show agreement with previous results with respect to second-
- generation upgrading [37] [77] [78] and third-generation upgrading of biogas [15] [16] [41] [46].

In equation 5, the capital charge factor (CCF) is equal to 0.08 [79]. Moreover, in equation 6 the total installation costs (TIC), indirect costs (IC) and owner's and contingencies costs (C&OC) are presumed to be 0.65, 0.14 and 0.15 respectively [17] [41]. The O&M fixed, O&M variable and equipment costs are calculated based on the assumptions in table 6. Ultimately, the LCOE is derived from the levelised cost of production and altered based on the energetic content per volumetric unit.

The assumptions in table 6 are derived from producer data, literature sources, quotas and reports. In this respect, the biogas upgrading occurs in a containerised solution that includes the biogas pretreatment step. Moreover, the bio-hydrogen production containerised solution contains the bio-hydrogen purification step by design. On top of that, the CCUS integration unit is presumed to have a fixed price per hydrogen output unit [80]. Moreover, the process data are obtained from the respective producers. Other data was presumed similar to literature sources and reports. Finally, the relevant data points are scaled based on a 5.5 MW biogas installation, which mimics large-scale average biogas production in the Netherlands [64].

Nonetheless, the bio-hydrogen unit cost price and LCOH are based on static model assumptions. Figure 4 shows the sensitivity of the LCOH over the key parameters of the economic

Component	Unit	Value
Feedstock		
Biogas price	€/ $Nm^3$	0.25
Pretreatment plant cost	€	0
Biogas upgrading		
Plant cost	€/( $Nm^3$ /h biogas)	conf.
Scaling factor	Green gas flow	conf.
Electricity consumption	kWh/Nm3 biogas	conf.
Reforming plant		
Plant cost	€/( $Nm^3$ /h hydrogen)	conf.
Scaling factor	Green gas flow	0.6
Electricity consumption	kW <sub>e</sub>	29.5
Water consumption	L/h	300
Cooling water	$m^3/h$	30
Compressed air	Nm <sup>3</sup> /h	4.5
CCUS integration		
CAPEX addition	€/kg H <sub>2</sub>	0.33
OPEX addition	$\epsilon/kg H_2$	0.17
Production	_	
Plant availability	% h/year	95
Extra total plant costs	% TPC	5
Other CAPEX	% IC	1
Lifetime	year	20
Interest percentage	% IC	5
Leverage percentage	% IC	50
Maintenance costs	% IC	2.5
Labour cost	€/FTE/year	46,800
Labour	FTE/year	0.5
R&D cost	% labour	20
Overhead cost	% labour	50
Electricity cost	€/kWh	0.10
Process		
$CH_4$ conversion	%	0.9
CO conversion	%	0.95
$H_2$ capture rate	%	0.85
$CH_4$ combustion	%	100
Capture rate reforming	$\% CO_2$	90
Capture rate fuel gas	$\% CO_2$	90
Capture rate upgrading	% <i>CO</i> <sub>2</sub>	100
Slip biogas upgrading	% CH <sub>4</sub>	conf.
Slip biogas upgrading	% CO <sub>2</sub>	conf.
Biogas composition	$v\% CH_4$	conf.
Biogas composition	$v\% CO_2$	conf.
Green gas composition	v% CH4 v% CO2	conf. conf.
Green gas composition	v /0 C O2	conj.

Table 6: Overview of relevant production, process and cost components for a 5.5 MW biogas installation used to determine the economical perspective of the  $_{625}$  different generation of biogas upgrading.



Figure 4: Sensitivity analysis of the LCOH for traditional steam green gas reforming for different levels of feedstock costs, CAPEX and OPEX

model. In figure 4 it can be seen that alterations in the feedstock price and or yield has the strongest effect on the LCOH. This can be explained by the strong contribution of the biogas feedstock costs to the total bio-hydrogen costs. This relates to the biogas input of  $9 Nm^3$  biogas/kg  $H_2$ , or around  $2.25 \notin$ kg  $H_2$ . In similar terms, an alteration in CAPEX, via the investment costs (IC) and labour costs, has more impact than changes in OPEX. This can be explained by the relative contribution of the different factors to the LCOH. Overall, the average LCOH can be stated to be in the range of  $4.47-5.35 \notin$ kg  $H_2$ . In similar terms, the average bio-hydrogen unit price is presumed in the range of  $3.75-4.47 \notin$ kg  $H_2$ . In case of biogas ATR, this reduces to  $4.02-4.80 \notin$ kg  $H_2$  and  $3.48-4.14 \notin$ kg  $H_2$  respectively.

Firstly, the variation in feedstock costs could be explained as a result of a reduction in biogas production costs and or biogas input. The biogas production costs are presumed to decrease due to an increase in production scale, commercialisation, technological efficiency, system optimisation and or favourable regulatory economic support [30] [81]. The biogas production costs could be reduced by 35%-45% in 2030-2050 by as compared to the 2018 cost levels [25] [78]. Moreover, improvements in the production process, like conversion, yield and capture rates could lower the feedstock demand [7] [66]. Secondly, with respect to CAPEX variations, the presumed production cost of biogas upgrading is assumed to lower with 10-20% as a result of economies of scale and technological improvements [37]. Additionally, the bio-hydrogen production CAPEX are presumed to decrease due to economies of scale, economies of scope, technological improvements and or process intensification options [37]. Lastly, In case of OPEX variations, improvements in industrial symbiosis, renewables integration, process and operations alterations and or efficiency improvements could support an overall reduction in OPEX [37]. Overall, these improvements could lower the apparent LCOH and bio-hydrogen unit cost to 3-5  $\in$ /kg  $H_2$ .

The bio-hydrogen cost structure also needs to be considered in relation to competitive hydrogen production methods to facilitate adoption of the concept of third-generation upgrading. In this respect, electrolysis technology is expected to see a sig-

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- nificant decrease in production costs over time and space [6]
   [7]. However, a reduction in the wider value chain costs could improve the relative retail cost position of bio-hydrogen. This relates to the significant contribution of the delivery costs in the overall cost perspective of hydrogen [66]. In this perspective,
- an additional 0.5-1.5 €/kg H<sub>2</sub> is presumed to be added to the bio-hydrogen production costs for on-site, local and or regional conversion, transportation and storage of bio-hydrogen. This is in contrast to an additional 1-6 €/kg H<sub>2</sub> in case of international transport or 1-3 €/kg H<sub>2</sub> in case of regional transport
  of hydrogen [7] [13] [38] [66]. In case of bio-carbon dioxide transport the additional cost could add around 30-65 €/t CO<sub>2</sub>
- for liquefaction or compression and transportation [82]. Therefore, the wider value chain costs restate the importance of local system optimisation and integration to improve the relative cost perspective of bio-hydrogen and facilitate the adoption of the
- concept of third-generation upgrading.

Moreover, the additional benefits associated with biohydrogen production support the relative cost position as compared to competitive hydrogen production methods. In case<sup>690</sup> of biogas production, these benefits relate to, for example, the sales of the by-product digestate that could stimulate an additional price benefit [26] [83]. On top of that, favourable regulatory support schemes and infrastructural developments could boost the relative price perspective [37] [64] [83]. Most impor-<sup>695</sup> tantly, the additional value of  $CO_2$  savings and or sales could improve the economic model [84]. This relates to the biocarbon dioxide yield of around 15 kg  $CO_2/kg H_2$  or 1.6 kg  $CO_2/Nm^3$  biogas. As a result, an increase in the bio-carbon dioxide value of 10  $\in/t CO_2$  would effectively reduce the bio-

- <sup>660</sup> hydrogen production cost price with 0.15 €/kg H<sub>2</sub>. However, based on a fixed carbon capture technology cost of 0.5 €/kg H<sub>2</sub>, and the observation that around 10 kg CO<sub>2</sub>/kg H<sub>2</sub> could be captured in the reforming step, an inherent bio-carbon dioxide value of 50 €/t CO<sub>2</sub> is required to recover the costs of the <sup>705</sup>
  <sup>665</sup> CCUS unit. As a result, the effective bio-hydrogen production costs price, as a result of the inclusion of a bio-carbon dioxide value of 80 €/t CO<sub>2</sub> [85], would reduce the bio-hydrogen
- production price point with 1.2  $\in$ /kg  $H_2$ , or 0.7  $\in$ /kg after the CCUS costs, to 3.2-3.7  $\in$ /kg  $H_2$ .
- Therefore, the concept of third-generation upgrading shows a higher cost perspective and lower energetic value, as compared to previous generations of biogas upgrading. This indi-715 cates the importance of the appropriate valuation of the additional bio-carbon dioxide stream. Moreover, additional benefits
- and lower relative value chain costs could further improve the price point of bio-hydrogen production. In this respect, system integration optimisation could stimulate the relevant cost<sub>720</sub> position of bio-hydrogen and bio-carbon dioxide. Overall, adequate valuation of bio-carbon dioxide shows strong potential
  to enhance the relative cost perspective. As a result, the conserver of the optimisation could appreciate a strong bio-production.
- cept of third-generation upgrading is supported over alternative biogas utilisation and competitive hydrogen production through<sub>725</sub> appropriate valuation of the bio-carbon dioxide.

$H_2$ value	$CO_2$ value $(\in/t CO_2)$	NPV	ROI (%)	Payback
$(\in/\ker H_2)$	( 1 2)	$(\in/\text{kg }H_2)$	0	(years)
3.6	80	-0.37	9	12
1.2		-25.79	-22	N/A
2.4	80	-13.08	-7	N/A
4.8		12.34	25	5
	40	-6.69	1	N/A
3.6	120	5.95	17	6
	160	12.28	25	5

Table 7: Investment parameters for traditional steam green gas reforming layout based on similar cost structure for 5.5MW biogas installation capacity and cost of capital of 7%

### 4. Discussion

The results indicate that the concept of third-generation upgrading is supported via a technologically feasible, environmentally benign and economically sound production process. Nonetheless, adequate valuation of the bio-carbon dioxide stream was indicated to be crucial for the adoption of the concept of third-generation upgrading.

Table 7 shows the net present value (NPV) investment parameter for the traditional steam green gas reforming layout based on the bio-hydrogen and bio-carbon dioxide value and a cost of capital of 7% [6] [86]. In table 7 it can be seen that the investment decision for the concept of third-generation upgrading is not supported at a bio-hydrogen value of 3.6  $\in$ /kg  $H_2$  [76] and a bio-carbon dioxide value of 80 €/t CO<sub>2</sub> [85]. Nonetheless, a small increase in the respective output value results in a positive investment decision. In this respect, the concept of thirdgeneration upgrading is supported at a bio-hydrogen price of 3.6 €/kg  $H_2$  and a bio-carbon dioxide price of 80-90 €/t  $CO_2$  or a bio-hydrogen price of 3.65  $\in$ /kg  $H_2$  and a bio-carbon dioxide price of 80  $\in$ /t CO<sub>2</sub>. This could be reduced to a bio-hydrogen price of 3.6  $\in$ /kg  $H_2$  and a bio-carbon dioxide price of 40-50 €/t  $CO_2$  or a bio-hydrogen price of 3.1-3.2 €/kg  $H_2$  and a biocarbon dioxide price of 80  $\in$ /t CO<sub>2</sub> for the biogas ATR layout. This reduces the required bio-hydrogen value by around 20% and the bio-carbon dioxide value by approximately 50%. Overall, the concept of third-generation upgrading is supported at a bio-carbon dioxide contribution of 25-30%.

Nonetheless, the concept of third-generation upgrading has to be considered within the context of competitive hydrogen production and alternative biogas upgrading generations. In this respect, the bio-hydrogen value is determined based on the available competitive production capacity. Moreover, the biocarbon dioxide value is determined in the context of negative carbon emissions and physical bio-carbon dioxide delivery. Table 8 shows the relevant price assumptions to determine the economic feasibility of the concept of third-generation over time. Here, the presumed total bio-hydrogen income in 2022 overlaps with the current hydrogen market income of 4-6  $\in$ /kg  $H_2$  [7]. Moreover, in table 8 it can be observed that the bio-hydrogen income reduces to 3.71  $\in$ /kg  $H_2$  by 2050. On the other hand, the total bio-carbon dioxide value, through physical sales of biocarbon dioxide and inherent carbon savings, increases from 115 €/t  $CO_2$  in 2022 to 400 €/t  $CO_2$  by 2050.

Income	2022	2030	2050	_
Sales value	2.20 [87]	3.60 [76]	2.00 [13]	-
(€/kg <i>H</i> <sub>2</sub> )				76
Subsidy value	2.46 <sup>a</sup> [72]	$0.21^{b}$	$0^c$	
(€/kg <i>H</i> <sub>2</sub> )				
Sales value	35 [82]	60 [82]	$200^{d}$	
(€/t <i>CO</i> <sub>2</sub> )				
ETS price	80 [85]	135 [88]	200 [89]	
(€/t <i>CO</i> <sub>2</sub> )				76
Sales value	1.5 [90]	1.1 [91]	0.30 <sup>e</sup>	
$(\in/Nm^3 \text{ GG})$				
				_

Table 8: Price assumptions of main output products over time period 2022-2050. <sup>*a*</sup> subsidy scheme similar to current green gas SDE+ subsidy. <sup>*b*</sup> Reflect difference in production cost and sales value. <sup>*c*</sup> Reflect no subsidy need by 2030-2050. <sup>*d*</sup> Reflect ETS trading price by assumption. <sup>*e*</sup> Reflect bio-hydrogen sales value by assumption.

	2022	2030	2040	2050	
NPV 3rd generation	20.76	26.94	33.41	38.86	
(€/kg <i>H</i> <sub>2</sub> )					
NPV 2nd generation	60.13	35.02	15.62	11.06	
(€/kg <i>H</i> <sub>2</sub> )					775
$\Delta$ NPV (3rd - 2nd)	-39.37	-8.08	17.79	27.80	

Table 9: NPV calculation for the 3rd and 2nd generation biogas upgrading over time period 2022-2050 in  $\notin$ /kg  $H_2$ 

Table 9 indicates that the concept of third-generation upgrading is supported over second-generation upgrading after 2030 and shows significant better economics by 2050. More specifically, the concept of third-generation upgrading is supported as of 2032 at a net bio-carbon dioxide value of around  $200 \notin t^{785}$  $CO_2$  and a bio-hydrogen net value of around  $3.5 \notin kg H_2$ .

Figure 5 shows that the bio-carbon dioxide value will contribute around 75% to the total income of the concept of third-generation upgrading over time. It is expected that the bio-carbon dioxide value will contribute over 50% to the total value<sup>780</sup> around 2035. This results from a decreasing value of bio-hydrogen, as a result of the commodity value of cheap renewable hydrogen, and following an increase in the value of bio-carbon dioxide, due to the need for emission reductions and demand for scarce bio-carbon dioxide in a climate neutral energy system. As a result, it can be observed that the concept of third-generation upgrading is dependent on the value of bio-rabon dioxide over time. This result from the value of nega-

tive emissions or circular usage of bio-carbon dioxide and the demand for scarce renewable carbon-containing molecules in an electron dominated energy system. Therefore, the concept of third-generation upgrading portrays the highest valorisation<sup>800</sup> potential of biogas through increased utilisation and valuation
 of the inherent bio-carbon dioxide over time.

Therefore, to support the renewed perspective on the upgrading of biogas, within the regulatory vision on a fossil free energy system, adequate boundary conditions should be in place.<sup>805</sup> This relates to the cascading principle of biogas, where the

highest societal value is achieved in applications with limited practical and or economic feasible alternatives. In this respect, the decoupling of the energetic bio-hydrogen and molecular bio-carbon dioxide supports the interpretation of the valorisation potential of the concept of third-generation upgrading from bio-hydrogen to bio-hydrogen and bio-carbon dioxide to ultimately bio-carbon utilisation. This could be achieved via market creation, infrastructural development and financial support mechanisms to address the relevance of biogas within the future renewable hydrogen energy system from a bio-hydrogen, biocarbon, system integration and or energy security perspective.

# 4.1. Bio-hydrogen perspective

The societal benefits of bio-hydrogen production relate to the zero pollution hydrogen product, the system infrastructure optimisation and the value chain cost benefits. Moreover, biohydrogen production directly adds relevant renewable hydrogen production capacity and lowers the environmental impact of fossil based production methods. As a result, the production of bio-hydrogen requires adequate governmental support to facilitate supply-side and demand-side adoption.

In this respect, the dominant policy options to stimulate the development of renewable molecules in the Netherlands, the 'Stimuleringsmaatregeling Duurzame Energie' (SDE) and the 'Hernieuwbare Brandstof Eenheid' (HBE), should be altered to support the concept of third-generation upgrading. In case of the SDE subsidy, this relates to a limited long-term perspective on the future renewable hydrogen energy system. This translates into uneven support for short-term and large-scale projects, a narrowly defined concept of CO<sub>2</sub> emissions savings per cost unit and the inability to value negative carbon emissions. In case of the HBE support scheme, the inherent biocarbon in biogas is insufficiently valued. As a result, the HBE requirement limits the potential for zero pollution fuels, the valuation of negative carbon savings and the efficient allocation of scarce carbon-containing molecules. Therefore, alternative support schemes should be prescribed to support the concept of third-generation upgrading within the envisioned renewable hydrogen energy system.

#### 4.2. Bio-carbon perspective

The societal benefits of the bio-carbon perspective relate to the potential to facilitate negative  $CO_2$  emissions through circular utilisation of bio-carbon dioxide and the contribution of biogas as scarce carbon-containing molecule within a fossil free energy system. This highlights the valorisation potential of renewable carbon-containing molecules to operate as feedstock in the bio-economy and unlock the feedstock transition [25] [67].

In this respect, the boundary conditions should resolve the current market failures, as result of the presence of negative externalities in the current energy system, which limit the adoption of the concept of third-generation upgrading [77]. The market failures relate to shortcomings like the inadequate valuation of bio-carbon dioxide and or negative  $CO_2$  emissions. Moreover, this is also associated with the environmental valuation of zero pollution hydrogen. Additionally, this relates to the limited perspective on the efficient allocation of renewable



Figure 5: Income streams of the concept of third-generation upgrading over time from 2022 to 2050 and the relative contribution of the bio-carbon dioxide value to the total income value

<sup>810</sup> molecules. Therefore, the concept of third-generation upgrading should be supported via market creation and supportive policy mechanisms that correctly value bio-carbon dioxide over<sup>840</sup> time within the proposed renewable hydrogen energy system.

## 4.3. System integration perspective

The societal benefits of the system integration perspective relate to the cost benefits associated with limited infrastructure needs. This supports the relative price point of bio-hydrogen<sub>845</sub> and bio-carbon dioxide in relation to competitive hydrogen production methods and alternative biogas applications. As a result, adequate infrastructural planning and development is re-

quired to stimulate the concept of third-generation upgrading. In this respect, a infrastructural development plan could besso

developed that focuses on the professionalisation and commercialisation of the biogas industry. The design would support the integration of processes, incorporation of renewables and the coupling of sectors [30]. In the infrastructure design, the production of bio-hydrogen is central to support the demand from<sub>855</sub> the decentralised industrial, agricultural, transportation and or build environment sector. Moreover, the production locations

- should account for end demand of bio-carbon dioxide and focus on the increasing relevance of bio-carbon dioxide within the future renewable hydrogen system. On top of that, local op-860 timisation should ensure limited value chain related costs and or requirements. As a result, the design should link the current infrastructural network with the biogas potential, the green gas
- production locations and the presumed end demand. The objective is to optimise from a system cost perspective, within the

boundary conditions related to the transport requirements of the biogas feedstock. Therefore, the infrastructure design could facilitate the adoption of the concept of third-generation upgrading through the cost-effective connection of bio-hydrogen and bio-carbon dioxide production with end demand locations.

# 4.4. Energy security perspective

The societal benefits of the energy security perspective relate to the problematic waste streams, the short supply chains and the geopolitical vulnerabilities. This underwrites the support for the concept of third-generation upgrading as local and or regional bio-hydrogen production method.

In this respect, the technical potential of AD biogas production is around 5.1 bcm natural gas equivalent in the Netherlands [29]. However, the economic potential is limited to 1.1 bcm by 2030. This could be lowered as a result of a decrease in livestock [29]. In perspective, the approximate total current natural gas usage in the Netherlands is around 40 bcm, where the power sector is responsible for around 30%, the industrial sector for 25% and the build environment for 20% [92]. As a result, the technical AD biogas potential represent around 3% of the natural gas demand in the Netherlands. This translates into 10%, 12% and 14% for the power, industrial, build environment sector demand respectively. On the other hand, based on a theoretical stoichiometric conversion potential, the technical AD biogas potential could fulfill around 45% of the current hydrogen production, or around 55% of the SMR produced hydrogen, in the Netherlands [93]. Therefore, the concept of third-generation upgrading shows could facilitate energy security.

#### 5. Conclusion

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Radical alterations in the current energy system are re-<sup>925</sup> quired in light of the dramatic effects of human-induced climate change. The envisioned renewable hydrogen energy system supports the need for a climate neutral energy system in the European Union by 2050. In this perspective, biogas is of<sup>930</sup> vital importance for the local and or regional production of biohydrogen and bio-carbon dioxide. This renews the perspective on the cascading principle of biogas from source of renewable

- electricity, heat or fuel to biogas as platform molecule able to<sup>935</sup> couple the renewable hydrogen and bio-economy domain. In this way, biogas is a relevant source of zero pollution hydrogen and an indispensable source of bio-carbon dioxide, which is presumed to become increasingly relevant in light of scarce re-<sup>940</sup>
- newable carbon-containing molecules in an electron dominated energy system. Therefore, the concept of third-generation upgrading renews the perspective on the valorisation potential of biogas and values the bio-carbon dioxide content in light of re-<sup>945</sup> quired negative carbon dioxide emissions and the need for, the
- circular utilisation of, renewable carbon-containing molecules in a fossil free energy system.

The concept of third-generation upgrading relies on a tech-<sup>950</sup> nologically feasible, environmentally benign and economically sound production process. In this respect, the adequate valua-

- tion and utilisation of the additional bio-carbon dioxide ensures a positive economic valuation of the concept of third-generation upgrading, despite a decrease in energetic value and increase in the relative price point. This is supported by the increased valuation and contribution of the bio-carbon dioxide value as 900
- compared to the bio-hydrogen value over time. As a result, biogas is initially valued as source of bio-hydrogen, later as source of both bio-hydrogen and bio-carbon dioxide and ultimately as source of bio-carbon.

Therefore, the concept of third-generation upgrading should<sup>300</sup> be supported via regulatory commitment, market creation, policy mechanisms and infrastructural changes. In this way, biogas could act as an invaluable source of bio-hydrogen and bio-<sub>970</sub> carbon dioxide, spark system integration and stimulate energy security in light of the renewable hydrogen energy system.

<sup>905</sup> Ultimately, the way biogas is seen should be changed.

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