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Carbon Capture in Hydrogen Production - Review of Modelling Assumptions

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ABSTRACT

The emission footprint of blue hydrogen production varies in literature, resulting in opposing recommendations on its eligibility in a carbon-neutral energy system. Next to fugitive methane emissions and global warming potential, the assumptions regarding carbon capture (CC) can significantly influence results. This analysis reviews these assumptions of several recently published blue hydrogen studies and compares them to experiences with CC in practice. It is found that the impact of various CC modelling parameters on emission footprints requires using real-world data in emission accounting. It is necessary to establish a concise emission accounting methodology for CC to increase transparency for stakeholders on blue hydrogen emissions.

Keywords: Carbon capture, blue hydrogen, low carbon technologies, steam methane reforming, autothermal reforming

NONMENCLATURE

Abbreviations	
ATR	Autothermal Reforming
CC	Carbon Capture
CCS	Carbon Capture and Storage
CCU	Carbon Capture and Utilization
LCA	Life cycle assessment
MDEA	Methyl diethanolamine
SCL	Syngas chemical looping
SMR	Steam Methane Reforming
(V)PSA	(Vapor) pressure swing adsorption
WGS	Water gas shift reaction

1. INTRODUCTION

Hydrogen has gained increasing importance as a key decarbonisation lever for climate-neutral energy systems [1–4]. Several countries and regions have published hydrogen strategies (see e.g. [2,5]). The strategies differ in what they consider as clean hydrogen production methods. While some strategies only see green hydrogen from renewable electrolysis as long-term compatible with a climate-neutral energy system, others also consider blue hydrogen from methane reforming combined with carbon capture (CC). In reference [6], an overview of the positioning of national strategies on hydrogen production is provided. For a number of countries, such as USA or China, significant production volumes of fossil-based hydrogen are foreseen in the long run. The IEA projects that in 2070, 40% of global hydrogen production will be from fossil fuels with carbon capture and storage (CCS) [7]. On the other hand, IRENA foresees only a transitional role for low emission hydrogen and shifts the focus to green hydrogen [8]. The ambivalent view on blue/low-carbon hydrogen is also mirrored in recently published life cycle assessment (LCA) studies that compare the emission footprint of different hydrogen value chains. Some sources [9,10] see blue hydrogen has an important element in the scale up of the market to a global hydrogen economy. Other studies [6,11] consider it as non-compatible with a climate-neutral energy system due to its residual emission footprint. The lack of transparency around the actual climate impact of blue hydrogen is reflected in regulatory uncertainty regarding its consideration as a low-carbon fuel, which in turn leads to investor uncertainties. There is a risk of stranded assets, if blue hydrogen has only a transitional role, as indicated by some models [12]. Albeit, it has to be considered that CO_2 infrastructure is likely to be used for more than just one project and will play a part in the decarbonisation of the industry sector, e.g. in the cement industry [3]. At the same time, highly optimistic projections on blue hydrogen's climate impact could lead to fossil path dependency and procrastination in green hydrogen infrastructure development [11].

The differences in the climate impact of blue hydrogen can stem from a variety of factors. A review of the methodological choices in blue hydrogen environmental assessment has been carried out by reference [10]. It was found that important parameter choices are blue hydrogen production technology, upstream methane emissions and the choice of metric to quantify impacts.

The assumptions around carbon capture and subsequent storage (CCS) or use (CCU) can be another decisive factor. It is therefore of importance to review CCS/CCU assumptions and their underlying rationality in dedicated studies to understand their effect on hydrogen's climate impact calculations. Furthermore, as assumptions are often based on theoretically achievable technical characteristics, a comparison to observed CCS/CCU activities' performance in practice can shed light on the gap between projections and reality. The value chain of CCS/CCU consists of many elements that each can have an effect on the overall emission footprint of blue hydrogen (e.g. capture technology, placement of capture in production process, capture rate, CO_2-compression, transport, use or storage). Due to the magnitude of effects to study, we focus on the assumptions around the actual capture process within the hydrogen production process in this short paper. Assumptions on transport, storage or use are not assessed. The overall aim of this analysis is to investigate how CC is modelled in selected hydrogen LCA studies to shed light on the varying conclusions. The findings can add to the policy implications regarding the consideration of blue hydrogen as a low-carbon fuel. Insights on the impact of modelling choices of CC in LCA studies can further inform greenhouse gas (GHG) accounting regulations for hydrogen certification schemes.

2. FRAMEWORK AND METHODOLOGY

The study focuses on natural gas based hydrogen production due to its potentially large role in future energy systems. A range of studies included the modelling of CC's effect on the climate impact of blue hydrogen. For this analysis, studies have been selected with opposing recommendations for action regarding the use of blue hydrogen, published within the last five years. The studies comprise of a mix of peer-review and grey literature (Table 1). These are investigated for the role CC played in the study's conclusions. First, the influencing criteria in the modelling of CC are identified. It is then evaluated if and how the studies have modelled these criteria. In a subsequent step, the findings are evaluated in the light of current status of CC in practice, considering among other things existing and planned CC plants and - as far as possible - achieved capture rates.

3. REVIEW OF HYDROGEN CARBON CAPTURE MODELLING ASSUMPTIONS

In the review of the underlying studies, five factors of influence in CC modelling have been identified: the reforming process, CC technology, placement of CC unit in the process, capture rate and CC energy consumption. The factors are not independent, but interact with each other. Not all studies define all of the factors. The following sections discuss the influence factors in detail.

3.1. Reforming method

Steam methane reforming (SMR) is the most established process in hydrogen production today. Two other established natural gas based processes are partial oxidation and autothermal reforming (ATR). The latter can be considered as a combination of SMR and partial oxidation [12]. Although ATR is less common at the moment, it is projected to increase in importance due to its favorable combination with CC [9,10,13].ATR can use the heat produced in the exothermal oxidation for the endothermic reforming reaction which takes place at the same time [10,14]. ATR produces only one emission stream from which CO_2 has to be captured [10]. Therefore, ATR is proclaimed to achieve higher capture rates compared to SMR. In an SMR plant, approx. 60% of emissions are produced directly in the reformer, 40% are created through the burning of natural gas in a furnace to provide reaction heat [9]. The evaluated studies consider both reforming processes. In many cases, SMR is used as a base case and ATR is added to show how higher capture rates can be achieved. ATR is considered to be easier to operate in terms of temperature management and heat requirements. However, the need for an air separation unit to provide oxygen for the partial oxidation reaction drives up the capital investment cost of the technology [15,16]. Moreover, ATR produces less excess steam that can be used for electricity generation in co-generation plants (e.g. SMR: +11,1 MW_e, ATR: +5,6 MW_e per 300 MW of H_2 [9]). Although ATR does not require burning natural gas in an external furnace, its lower hydrogen-to-carbon ratio requires to burn more natural gas as feedstock (0.15 GJ/kg H_2 for ATR compared to 0.12 GJ/kg H_2 for SMR) [14].

3.2. CC technology

Different methods and materials exist to perform the separation of CO_2 from the output streams. CO_2 separation in hydrogen synthesis can be considered as a form of hydrogen purification, which is a standard element in fossil fuel based hydrogen production. Therefore, synergies between the purification device (usually a pressure-swing-adsorption unit) and the carbon capture can be used.

The studies specifically mentioning the CC technology use mostly the state-of-the-art amine-based chemical absorption. Other cited technologies include vapor pressure swing adsorption (VPSA), membrane and

Table 1: Overview of assumptions taken by blue hydrogen studies regarding the modelling of CC (N.d. refers to "not disclosed"). As many of the studies evaluate several cases, the range of results is given. For case-specific results, refer to the indicated source. ATR = Autothermal reforming, CLR = Chemical looping reforming, MDEA = Methyl diethanolamine, NG= Natural Gas, SCL =Syngas chemical looping, SMR = Steam methane reforming, (V)PSA = (Vapour) pressure swing adsorption, WGS = Water gas shift reaction.

Study	H2 prod.	CCS technology	Placement of CCS in process	Capture rate	Process efficiency	Energy penalty of CC	Emissions in kg CO_{2eq}./kg H_2, (of which CC)
[6]	SMR	N.d.	Syngas	56-90%	78%	4% - 9%	3.4 - 7
[9]	SMR, ATR	VPSA/MDEA (amines)	PSA/WGS syngas (pre-combustion)	55-98%	77-78%	5 to 11 MW_e reduction in elec. surplus per 300 MW of H_2[1]	2.6 - 5.8
[10]	SMR, ATR	MDEA	WGS Syngas	55-93%	76-77%	N.d.	Approx. 3 - 6[2]
[11]	SMR	N.d.	Process + flue gas	Flue: 65% Process: 85%	78%	8% - 15% (+ 25 - 39% emissions)	16.2 - 16.7 (2.1 - 4.1)
[14]	SMR, ATR	Amines	WGS syngas	52-85%	67-80%	12% - 19% NG (SMR), +3 kWh/kg H_2 (ATR)	3.9-8.2
[15]	SMR, ATR, SCL, CRL	Amines	WGS syngas/flue/ chemical looping gasification	90-100%	65%-85%	1% - 18%	0-0.9
[17]	SMR	Amines	Syngas or PSA tail gas	90%	76%-85%	11% - 15%[3]	4.7
[18]	SMR	N.d.	Not specified	70-90%	60-85%	5% - 14%	13 (2.5)
[19]	SMR	Amines	Process + stack gases	90%	>75%	N.d.	3 - 9.2
[20]	SMR	MDEA, cryogenic, membrane separation	WGS syngas, PSA tailgas, flue gas	53-90%	76%	Up to +10% NG & 10 MWe reduction in elec. surplus per 300 MW of H_2	1 - 4.3

[1] The energy consumption of CC is calculated via the electricity balance of the plant.

[2] Results for the case with Global Warming Potential of 100 and a CH_4 emission rate of 1.5%.

[3] Reduces plant efficiency to 65-70%.

cryogenic separation [21], although these are less mature. The choice of technology has an influence on where the separation unit can be placed in the hydrogen production chain (section 3.3). Influencing factors are whether the capture is placed in a new or a retrofitted plant, the CO_2-concentration in the output stream, pressure and fuel type [22]. According to reference [23], amine-based absorption technology fits best to output streams for the reformer, cryogenic solutions to the output of the water gas shift and membranes to the output of the PSA unit. In reference [9], it is shown that the CC energy consumption in relation to capture rate differs based on the applied technology. They compare solvent based Methyl diethanolamine (MDEA) and VPSA. MDEA shows an exponential increase of energy consumption when capture rates exceed 97%. For VPSA, the additional energy consumption depends on the syngas purity, but the technology generally consumes more electricity than MDEA (-11.3 MW_e compared to -6.5 MW_e for the ATR 98% capture case per 300 MW of H_2). The sensitivity of the employed capture technology to energy requirements is an important observation for plant design and necessary specifications for GHG accounting methodologies. Blue hydrogen LCAs should also include an environmental assessment of the CC technology. LCA studies rarely include the emission balance of the capital goods in the supply chain, but it is recommended to do so [10]. The different CC technologies' footprint should also be included to receive a more complete picture of the benefits of each technology for the supply chain emission impact.

3.3. Placement of CC unit in process

Depending on the natural gas reforming process and the CC technology, the CC unit can be placed in different locations. The terms slightly differ in the studies for the locations of the CC unit [9,20]. Pre-combustion CC generally refers to capturing the emissions from the syngas or the PSA tail-gas, while post-combustion refers to the capture of emissions from burning the e.g. PSA tail-gas and additional natural gas in a furnace to provide heat [9]. This is often also referred to as flue gas capture

[11]. With the aim to achieve high overall emission reductions, the trend goes towards placing CC to capture plant-wide emissions in SMR, i.e. from the water-gas-shift reaction output syngas and from flue gas [9]. However, according to reference [20], using only pre-combustion CC on the syngas output is more economic and current practice. Therefore, if the CC is extended to capture plant-wide emission, the additional cost has to be quantified for blue hydrogen cost [6]. According to IEAGHG, costs are increased by 18% with a 56% capture rate and by 79% with a 90% capture rate [21].

3.4. Carbon capture rate

The carbon capture rate for natural gas based hydrogen production ranges from 53% to 100% in literature. 100% is assumed in reference [15], for chemical looping reforming processes, a process that is still in development. The large range of capture rates is a result of the selected reforming processes and the placement of the CC unit in the process [11]. The evaluated studies often compare a low and a high capture rate to show what is current practice and what is (theoretical) technologically feasible. 90% capture rate seems to have emerged as a standard high capture rate value in literature, as can be seen in Table 1 and according to reference [24]. This is also reflected in the CertifHy hydrogen certificate for low-carbon fuels, which will likely foresee a 60% reduction in greenhouse gases in relation to a fossil comparator. To achieve this, a 90% overall capture rate is necessary according to reference [6]. It has to be kept in mind that although higher capture rates can technically be achieved, the associated technological requirements could increase the cost of blue hydrogen [7] and reduce its (current) economic advantage over green hydrogen.

3.5. CC energy consumption

CC requires energy, i.e. steam for solvent regeneration and electricity for CO_2 compression [10]. In the underlying studies, the energy penalty of CC is mostly regarded as a reduction in efficiency of the overall process. It is assumed that the energy requirements for CC are served by burning more natural gas to produce electricity. Some studies expect that the additional energy needs for CC can be supplied within the reforming process, i.e. from exothermal reaction heat, and the energy penalty is not explicitly stated. E.g. reference [10] assumes that adding CC leads to only a small reduction in overall plant efficiency, requiring no or only little additional natural gas. This however requires to design the reforming process and the capture unit in an integrated way. In reference [9], the power and steam for the capture unit is generated in an integrated co-generation plant. Yet, it has to be considered that reforming processes are often net-exporters of steam e.g. for electricity production, which is then reduced and potentially lacking in other connected processes [21]. E.g. in the case of reference [9], the SMR process produces 12 MW_e of electricity in the co-generation unit, which is reduced to 3.5 MW_e with MDEA solvent and 0.5 MW_e with VPSA capture per 300 MW of H_2.

As the process efficiency of natural gas reforming itself differs from study to study, also the overall efficiency after subtracting CC differs. The reforming process efficiency in the investigated studies ranges between 60% to 86%. The energy penalty for CC varies significantly. One explanation for the variation is that higher capture rates lead to higher energy consumption. E.g. in reference [6] the low energy penalty of 4% is from a case of capturing only the process stream with 56% capture rate, while the high 9% energy penalty is a result of extending the capture rate to 90%. However, when looking at the same level of capture rate, the studies still exhibit a large range of values for energy consumption. This demonstrates the influence of differing modelling assumptions for plant full load hours, reforming temperature and pressure, technology and placement of CC, efficiency of steam and of electricity generation as well as level of CO_2 compression and drying [14,20]. The emissions of the entire plant and the CC unit are greatly influenced by the level of integration and resulting use of by-products, e.g. the availability of excess low grade heat for steam generation for the amine units [14]. Some studies only report the electricity consumption of the CC, potentially assuming the electricity source can either come from an integrated co-generation plant or from an external source. It is often advocated to use renewable electricity to power the CC unit, to not add additional emissions to the process. However, using renewable electricity for hydrogen production is a complex matter in itself, as illustrated by the current discussion on additionality criteria for electricity used in hydrogen production (see Delegated Act on Article 27 of the Renewable Energy Directive II [25]).

4. CURRENT STATUS OF CC IN PRACTISE

4.1. CC plants in operation

In September 2021, around 27 CCS plants were operational with a capacity of around 37 Mt p.a. [30]. However, the majority of CC is employed in natural gas processing, where different technological requirements apply, compared to its use in hydrogen production. In general, pairing hydrogen production with CC is regarded

as an efficient process due to the concentrated CO_2 output stream [6]. Only eight hydrogen production plants equipped with CC exist today, of which four are methane reformation based hydrogen producers. These are plants from Enid Fertilizer, Air Products (both USA) as well as Quest (Shell) and Alberta Carbon Trunk Line (both Canada) [26–29]. As of 2020, the total fossil hydrogen production with CC was 0.4 Mt and captures around 6 Mt of CO_2 [7]. Only 1% of global hydrogen production is blue hydrogen [26]. Reported data however varies due to divergences between planned project capacities, project timeframes and actual plant performance. Many plants employ CC to use the CO_2 for enhanced oil recovery. Here the CO_2 is not captured permanently, but to a large extent (70%) reemitted into the atmosphere. Real capture rates are rarely reported [6]. Theoretical assumptions are deemed too optimistic and demonstration projects are often delayed or did not meet expectations [19]. Although theoretical capture rates of over 90% are often cited, currently operating ones usually only achieve 50% - 60% plant-wide capture rates [10]. The Alberta Carbon Trunk line is projected to capture up to 14.6 Mt [28], current rates are in the order of 1.3-1.6 Mt [29]. According to the IEA Sustainable Development Scenario [7], 601 Mtoe of fossil hydrogen production with CCUS require to capture and store 1,900 Mt of CO_2.

4.2. CC project outlook

According to reference [29], the CCS project pipeline amounted to an annual capacity of 111 Mt CO_2 capture in September 2021. Of that capacity only a small amount of 3.1 Mt is labeled as in construction. Those projects have a reported operation date between 2021 and 2024. None of the four projects, however, directly relates to hydrogen production. Of the projects with a facility status labelled as advanced development (46.7 Mt capture capacity per annum), in total only 5 projects focus on the production of hydrogen. Of those five projects only two so far list a capacity amounting to 1.7 to 2.2 Mt p.a.. All five projects are developed within Europe, four of them are located in the Netherlands with the starting year indicated to be 2024, one of them is located in the UK with the starting year indicated to be 2028. The majority of CCS capacity (60.9 Mt) is labelled as early development with reported operation dates mainly laying between 2025 and 2030. Within that group, nine projects focus on the production of hydrogen of which seven report a capacity of in total 6.6 to 7.8 Mt storage per annum. Of those nine projects, five are located within the UK, one in the US, Sweden, Italy and Canada each.

5. EVALUATION

5.1. Evaluation of modelling assumptions

The evaluated studies' assumptions around key CC performance parameters show a bandwidth which influences the outcome of blue hydrogen having a higher or lower climate impact. Using ATR allows to assume higher capture rates without a high CC cost penalty because the process does not require capturing flue gases. Assuming a low energy penalty (no additional natural gas or no external energy source) will lead to no or low additional emissions. That is, if the indirect effects of reducing the process' net energy export are not included. The last 5% to 10% of capture are feasible at additional cost and energy requirements, but are usually not addressed, as most studies' highest capture rate is 90% [24]. ATR is proclaimed to be more suitable for CC, but it plays a minor role in hydrogen production at the moment [14]. Economic evaluations exist mostly for SMR so far. ATR capital cost may be higher than SMR due to the requirement of an air separation unit, but could be lower in OPEX due to lower steam use. Investment cost will depend on whether an existing reforming plant is retrofitted with CC, or if a new plant is planned with integrated CC. Retrofitting can make use of existing infrastructure, but new plants will likely be more efficient due to their level of integration of energy flows.

However, new plants will have a longer payback period than retrofitting, which contradicts a transitional role of blue hydrogen. SMR retrofitted with CC has been assessed in literature [17], but there are too few comparative assessments to derive general conclusions. The large uncertainty in SMR and ATR cost has to be further evaluated due to its implications for the cost advantage of blue hydrogen [14]. Looking at the evaluated studies, high blue hydrogen emissions e.g. from reference [11] assume that adding CC adds 2.1 kg $CO_{2\text{-eq.}}$/kg H_2 for CC without flue gas capture and 4.1 kg $CO_{2\text{-eq.}}$/kg H_2 with flue gas capture. The study assumes relatively low capture rates (65% -85%) and a high energy penalty that adds emissions of 25% - 39%. Lower blue hydrogen emissions, e.g. in study [9], comprise of assumptions of high capture rates of up to 98%, with novel capture technologies (VPSA) and with the energy penalty calculated as a reduction in electricity export. As our analysis shows, there is neither a standard CC process for hydrogen nor sufficient experience with the real-world application of CC in hydrogen production. The actual operation of the reforming plant is a deciding factor in the overall performance of CC in blue hydrogen production. This includes plant full load hours, CC technology and placement, level of process integration,

reforming temperature and pressure, efficiency of steam and of electricity generation as well as level of CO_2 compression and drying. It is therefore of high importance to use real plant data in emission accounting. The use of default emission values, which are often employed e.g. in certification of fuels to limit the administrative burden, risks to cover up poorer plant performance and can result in a significant overestimation of emission reductions achieved through CC. Reference [30] criticizes that each study creates their own accounting framework, setting system boundaries in different places, which affects the results. A common approach (e.g. in the form of guidelines from an international organization similar to GHG reporting guidelines from the IPCC) could help to overcome that shortcoming.

5.2. Implications for CC infrastructure

A key advantage of blue over green hydrogen is its lower production cost, which are also a consequence of being able to use existing infrastructure. If blue hydrogen is produced from SMR, large capacities already exist today that could be equipped with CC [17]. Using existing SMR infrastructure would require to capture both process and flue gas emissions, as otherwise blue hydrogen's emission would be too high to be classified as a low carbon fuel. This could potentially pose a significant economic and technical difficulty on the plants, as existing SMR plants are often highly integrated with other processes (e.g. to export excess steam). These energy exports would have to be replaced. As a result, existing cost benefits compared to green hydrogen are likely to be diminished. The necessary high capture rates needed for sufficient emission reductions in the longer-term are likely only achieved with ATR. The extent of retrofit or new installations necessary has a significant impact on the infrastructure investments that are necessary for scaling up the production volumes. According to the IEA, ATR is already in use by a large share of ammonia and methanol production. However, for ATR hydrogen production with CC, only two planned demonstration projects in the UK are reported [7]. Cost additions for blue hydrogen due to use of ATR and the last 5% - 10% additional capture to reach rates close to 100% have to be quantified to provide a realistic cost estimate of blue hydrogen with a truly low emission footprint. The high infrastructure investments and operating costs for high capture rates combined with the rather slow and regionally quite focused development of blue hydrogen plants makes it more and more unrealistic, that significant investments into blue hydrogen will pay off as long as there are still significant amounts of emissions left.

6. CONCLUSIONS AND OUTLOOK

If renewable energy capacity developments continue to lag behind their expansion goals, blue hydrogen is projected to be employed, at least for an interim period, to reduce the accumulation of GHG in the atmosphere in the years leading up to 2050. However, our analysis shows that the emission benefit of blue hydrogen also greatly depends on the CC process design. In most studies advocating the use of blue hydrogen, CC is depicted in a beneficial process configuration: using ATR instead of SMR, assuming a low energy penalty that can be supplied within the system. However, achieving higher capture rates - which are needed to realize significant emission reductions - will lead to a higher energy penalty from CC, offsetting potentially some of the emission savings.

The analysis shows that there is not a standard hydrogen CC process, which emphasizes the necessity to use real-world data for emission accounting, e.g. in a future hydrogen certification system. A concise and comprehensive emission accounting methodology should be established that prevents the risk of hidden embedded emissions due to different plant performance and reporting practices. The different factors of influence on the impact of CC in hydrogen production require future in-depth research. On the one hand, the impacts of the various CC technologies have to be included in blue hydrogen LCAs. On the other hand, the variations in energy input into CC have to be further investigated to quantify their effect on the overall emission balance. As long as there are still significant emissions involved in producing blue hydrogen it can only be seen as a bridging technology. However, the slow and regionally focused development of blue hydrogen production plants lowers the chances of such investments to pay off. Cost advantages of blue hydrogen compared to green hydrogen are sensitive to the cost of CC, which in turn rely on plant-specific performance, capture rate and the level of plant integration. The use of ATR plus CC for hydrogen production would require a high number of greenfield investments compared to retrofitting existing SMR plants with CC.

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