On the cost of zero residual CO_2 emission hydrogen: A techno-economic analysis of steam methane reforming with carbon capture and storage

Daniel Mullen[*a], Laura Herraiz[b], Jon Gibbins[c] and Mathieu Lucquiaud[a,c]

a School of Engineering, The University of Edinburgh, Edinburgh, United Kingdom
b School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh, United Kingdom
c School of Engineering, The University of Edinburgh, Edinburgh, United Kingdom

Abstract

In a net zero economy compliant with the emission targets implicit in the Paris Climate Change Agreement any residual greenhouse gas emissions from hydrogen produced via steam methane reforming (SMR) of natural gas will require reduction to zero.

The initial step to achieving this is the capture and storage of 100% of the fossil carbon from both the natural gas feedstock and the natural gas used for combustion, hereafter referred to as zero residual CO_2 emission production. For an SMR this means achieving an overall CO_2 capture fraction of 99.8% from the flue gases. The remaining 0.2% of stack CO_2 emissions are then equal to the atmospheric CO_2 from the combustion air used in the steam methane reformer furnace. Via the process modelling of a steam methane reformer integrated with post-combustion CO_2 capture (using the CCSI MEA steady state model [1]), we predict that, for an open-art solvent of 35%wt aqueous MEA, a capture fraction of 99.8% and a peak desorber temperature of 125°C, the CO_2 absorber column requires 20m of structured packing and the intercooling of the solvent, compared to an estimated 14m of structured packing and no intercooling for 5% residual emission hydrogen (95.2% capture fraction). For a 1 GW hydrogen production facility, we use public domain cost studies to estimate that this translates to an increase in total capital requirements of 6.5%, from £987M to £1051M, and a marginal increase in specific CO_2 capture energy requirements of 1.6%, from 3.62 GJ/tCO_2 to 3.67 GJ/tCO_2 of low grade thermal energy, provided by low pressure steam extracted from the process. The resulting net operating costs, including CO_2 transport and storage, natural gas fuel and steam for solvent regeneration but excluding any CO_2 emission costs associated with the cases with residual emissions, increase by 3.7% from £468M to £485M p/a. The additional CO_2 captured reduces hydrogen production efficiency by 1.9 percentage points on an HHV basis (from 68.7% HHV to 66.8% HHV).

For 2020 UK capital costs, an assumed natural gas price of 28 £/MWh$_{th}$ HHV (82 p/therm) and average electricity selling price of 90 £/MWh$_e$, hydrogen can be produced with zero residual emissions at a Levelised Cost of Hydrogen (LCOH) of the order of 62 £/MWh$_{th}$ HHV (2.4 £/kg H_2) with an associated levelised cost of capture (LCOC), excluding transport and storage, of 54.1 £/tCO_2. This translates to a marginal increase of 0.34 £/tCO_2 per percentage point increase in net capture fraction when compared to operating with 5% residual emissions (52.4 £/tCO_2). Hydrogen production with residual CO_2 emissions of 5% results in the same LCOH of 62 £/MWh$_{th}$ HHV with a carbon price of 172 £/tCO_2.

Keywords: Carbon Capture and Storage, Hydrogen, Net Zero, PCC, Ultra High CO_2 Capture

1   Introduction

The need for rapid scale up of low GHG emission hydrogen production has been reflected in the Sixth Carbon Budget [2], issued by the UK Committee on Climate Change, and the 2021 UK hydrogen strategy [3], which suggests that 250-460TWh HHV (Higher Heating Value) of hydrogen will be needed in the UK annually by 2050, making up 20-35% of total energy consumption. For comparison 27 TWh HHV of hydrogen was produced in the UK in 2016 [4], primarily for industrial processes such as ammonia production. The UK has recently set a target of installing up to 10GW of low carbon hydrogen production capacity by 2030 [5] in order to meet the goals set out in the Sixth Carbon Budget.

Large-scale low carbon hydrogen production in the UK is expected to be mainly accomplished by a combination of the following methods:

- Electrolysis using dedicated or excess renewable electricity (green hydrogen). Hydrogen produced in this fashion provides one route to low carbon hydrogen with a carbon intensity of 0.1 gCO_2e/MJ H_2 Lower Heating Value (LHV). However, currently Levelised Costs of Hydrogen (LCOH) in excess of 109£/MWh$_{th}$ HHV are reported by the UK Department for Business, Energy and Industrial Strategy (BEIS) due to high capital costs, low load factors and limited availability of surplus electricity [3, 6].
- Methane reforming with CO_2 capture (blue hydrogen) in a Steam Methane Reformer (SMR) or an Autothermal Reformer (ATR). This includes a Carbon Capture and Storage (CCS) system, which separates the CO_2 produced in the process for permanent geological storage. . The carbon intensity of the hydrogen produced varies depending on the CO_2 capture fraction applied. Reported values typically lie between 0 – 21.4 gCO_2e/MJ H_2 (LHV) [3]. In the past, lower capture fractions (~90%) have been assumed for SMR+CCS and higher values (~ 95%) for ATR+CCS (e.g. see [6]). In a recent UK study LCOH for SMR+CCS production with a natural gas price of 19-46£/MWh$_{th}$ HHV are reported at 39-73£/MWh$_{th}$ HHV with a scope 1 carbon intensity of 8.7 gCO_2e/MJ H_2 (LHV)[6].

As noted above, for blue hydrogen production the amount of CO_2 captured relative to the amount of CO_2 produced (CO_2 capture fraction) in steam methane reformers is often assumed to be 90% and, with the expected future increases in CO_2 emission costs, this will lead to significant escalation in the predicted cost of hydrogen production using SMR+CCS. It is clear that 10% residual emissions, if truly unavoidable, cast doubt on the viability of SMR+CCS produced hydrogen as a climate mitigation technology in the context of an energy system compatible with net zero ambitions. Recent UK guidance by the UK Environment Agency [7, 8] considers a 95% CO_2 capture fraction as the current Best Available Technology (BAT) for post-combustion CO_2 capture from biomass and gas-fired thermal power plants, while a 95% or above capture fraction for SMR+CCS is considered by Curtis et al. in a recent BAT review on hydrogen production methods [9]. The National Energy Technology Laboratory in a report for the US Department for Energy found that transitioning from an unabated SMR to operating with a 96% capture fraction increased the LCOH from 1.06 \$/kg H_2 to 1.64 \$/kg H_2 [10]. The UK low carbon hydrogen standard published in 2022 considers 20 gCO_2e/MJ H_2 (LHV) the maximum net carbon intensity (inclusive of all supply chain and construction emissions) threshold for acceptance as low carbon hydrogen [11].

There is now a growing body of evidence suggesting that ultra-high capture fractions, defined here as CO_2 capture fractions equal to or higher than 99%, can be technically and economical feasible [12-17]. Feron et al. reports that increasing the CO_2 capture fraction of a post-combustion solvent-based system (30 %wt monoethanolamine (MEA)) from 90% to a zero residual emission level of 99.7% would give a 1.5 percentage point reduction (34.5% to 33% LHV) in thermal efficiency on LHV basis for a ultra-supercritical coal fired power plant, and a 2.2 percentage point reduction for a natural gas fired combined cycle (48.6% to 46.4% LHV) [12]. Work completed by Danaci et al. reports that, for a representative flue gas flow rate of 500 kg/s, and flue gas CO_2 concentrations of 4% vol, 10% vol and 20% vol, transitioning from 90% to 99% CO_2 capture fraction with 30 %wt MEA results in an increase in total capture cost of 7%, 10% and 13% respectively [13]. A process modelling study of CO_2 post-combustion capture from a combined cycle gas turbine power plant by Michailos and Gibbins reported that transitioning to 99% CO_2 capture from 95% can be achieved with a moderate increase of 7.7% in the specific thermal energy input to the reboiler (specific reboiler duty) of the CO_2 capture process for 35 %wt MEA , providing the absorber is sufficiently sized and the stripper pressure is high enough to avoid excessive water vapour in the product CO_2 at the reduced lean loading required [14].

Pilot scale tests at the National Carbon Capture Center (NCCC) completed by Gao et al. find that increasing the CO_2 capture fraction of a coal-fired power plant from 90% to 99% resulted in an increase in specific reboiler duty of lower than 5% with a 5 m piperazine (PZ) solvent solution [15]. Tests completed at Technology Centre Mongstad show that, with 24 meters of packing, 35 - 37 %wt monoethanolamine (MEA) solvent and a CO_2 % vol of 3.9-4.2%, 99% CO_2 capture can be achieved with a specific reboiler duty of 3.8 GJ/tCO$_2$ compared to 3.6 GJ/tCO$_2$ for a 90% CO_2 capture fraction [16], approximately a 6% increase. Hirata et al. working with Mitsubishi Heavy Industries Engineering, Ltd using a proprietary solvent (KS-1™) investigated 99.5% CO_2 capture fraction for a reference 650 MW$_e$ coal fire power plant and predicted that near zero emissions could be achieved with a 3% increase in the total annualised cost of CO_2 Capture ($/tCO$_2$) [17].

This article builds on these previous studies to estimate that zero residual carbon emission H_2 production via SMR+CCS can be achieved with an overall plant CAPEX increase of 6.5%, an increase in specific reboiler duty of 1.6% and a 1.9% point reduction in H_2 production efficiency (HHV), relative to a 95% CO_2 capture reference case.
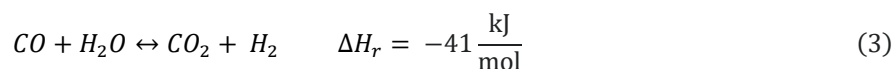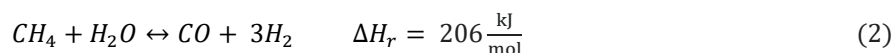
2 Methodology

2.1 *Modelling methodology for a Steam Methane Reformer & Post combustion CO_2 capture Plant*

A model of a 1000 MWh$_{th}$ HHV SMR is developed in gPROCESS Process Builder [18], a process modelling platform that allows the creation of bespoke unit models for each specific unit operation. The SMR process flow diagram is illustrated in Figure 1. Technical and operational parameters are based on a conventional SMR with post-combustion CO_2 capture presented in a report commissioned by the IEA Greenhouse Gas R&D Programme (IEAGHG) in 2017 [19].

The incoming ambient air is preheated to 150-170 °C using residual heat from the furnace exhaust flue gas. The preheated air then enters the burners where a mixture of natural gas and off-gases from the hydrogen production process is used to raise the flue gas temperature up to 1800-1900 °C, whereupon it passes over the reformer tubes to provide heat to an endothermic reforming reaction converting natural gas to hydrogen and carbon monoxide. The flue gas leaves the reformer at ~950 °C and the remaining heat is used to reheat process synthesis gas (syngas) and generate steam at 42 bar and 400°C for the reforming process, power generation and thermal energy input to the CO_2 capture plant for solvent regeneration. As noted above, the remaining low grade heat is then used to preheat the incoming ambient air before the flue gas is cooled prior to entry into the post-combustion CO_2 capture plant.

High pressure steam and preheated natural gas feedstock at 500°C and 34 bar are mixed and enter a pre-reformer, which is an adiabatic reactor that converts 100% of the C$_{2+}$ hydrocarbons and olefins present in the feedstock into CO and H_2. The ratio of steam to natural gas is controlled to give a steam to carbon ratio of 2.5 in the reformer. After pre-reforming the syngas is re-heated to 600°C prior to entry to the catalyst-filled reformer tubes where two concurrent reactions occur to form a syngas consisting of equilibrium proportions of CH_4, H_2O, H_2, CO_2 and CO at ca. 913°C. The Steam-Methane reforming reaction as described by the equilibrium shown in Equation 2 and the Water-Gas shift reaction as described by Equation 3. Less than 100% methane conversion in the reformer is expected due to process pressure and temperature limitations and a 85% methane conversion rate is calculated for the for the above process conditions; this unreacted methane is typically known as 'methane slip'.

$$CH_4 + H_2O \leftrightarrow CO + 3H_2 \qquad \Delta H_r = 206 \frac{\text{kJ}}{\text{mol}} \qquad (2)$$

$$CO + H_2O \leftrightarrow CO_2 + H_2 \qquad \Delta H_r = -41 \frac{\text{kJ}}{\text{mol}} \qquad (3)$$

This syngas is cooled to 320 °C via an evaporator prior to entry into the water gas shift reactor (WGSR), where further conversion of CO occurs (ca. 72%) via the water-gas shift reaction.

The remaining useful heat in the syngas stream is used to pre-heat the natural gas and the water condensate in the

power cycle. Final cooling and condensation of the syngas to 35°C is completed before entering a pressure swing absorber (PSA), wherein it is assumed that 90% of the H_2 [19] is separated out at 25 bar for export. The off gas, consisting of the remaining H_2, unreacted CH_4, CO and the CO_2, is mixed with additional natural gas for combustion in the burners of the SMR furnace. Due to the high grade heat requirement of the reforming process, the flue gas has useful energy in excess of that useable for natural gas reformation; this is employed to raise steam, some of which is fed to the reformer, with any surplus steam diverted to a steam turbine for power generation, or simply exported for off-plant use, depending on site specific conditions. In CCS applications, surplus steam would be typically be diverted to a back pressure turbine with an exhaust pressure suitable for the supply of thermal energy to the CO_2 capture plant at constant temperature, in this case 3.7 bar. For the capture fractions used in this study the thermal requirement of the CO_2 capture process is shown to be in excess of the low pressure steam available in an unabated SMR. As a result additional steam production is required, necessitating an increase in supplementary fuel and ambient air intake.

The CO_2 capture system in this study consists of a conventional chemical absorption system using a 35%wt monoethanolamine (MEA) aqueous solution, an open-art solvent for CO_2 capture processes. In principle any viable commercially available post-combustion CO_2 capture technology could be used, although it would have to be determined whether or not it is possible to achieve such high capture fractions. Process modelling for the design and optimisation of the capture plant is conducted using an open source model [20] developed by the Carbon Capture Simulation for Industry Impact (CCSI^2) partnership [1] in Aspen Plus [21], which was developed using pilot scale data provided by the US National Carbon Capture Centre (NCCC) [22]. Figure 1 shows the process flow diagram of the MEA based capture process as implemented in ASPEN/CCSI.
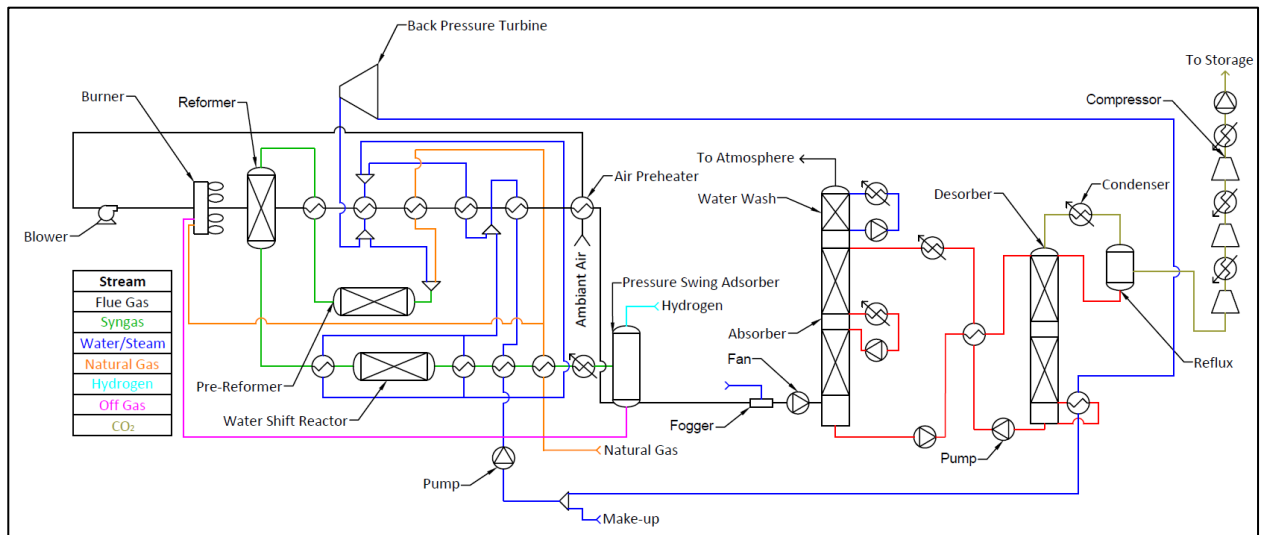

Figure 1: Steam methane reforming process with flue gas CO_2 capture and compression to storage

The flue gas exits the air preheater in Figure 1 and initially passes through a water fogger, which injects atomised cooling water, cooling the flue gas to saturated conditions at 40 °C. A booster fan then increases the flue gas pressure sufficiently to overcome the pressure drop through the absorber train. The flue gas enters the bottom of a counter current packed bed absorber while CO_2 lean MEA solvent (lean solvent) enters the top. The ratio of lean solvent to flue gas flow rate is often referred to as the liquid to gas ratio or L/G ratio and is set to achieve the required CO_2 capture fraction in the absorber, in conjunction with other process parameters. As the flue gas passes up through the solvent-laden packing CO_2 transfers to the liquid solvent and reacts exothermically, heating the flue gas and solvent. The CO_2 depleted flue gas leaves the top of the CO_2 capture packing and then goes through a water wash section, to remove residual MEA prior to release to the atmosphere, while the now CO_2 rich solvent (rich solvent) leaves the bottom of the absorber to be stripped of CO_2.

If the partial pressure of CO_2 in the flue gas was constant throughout the absorber, the driving force for CO_2 absorption decreases towards the top of the column as the solvent absorbs CO_2 and higher temperatures resulting in a decreased

mass transfer rate. When the CO_2 partial pressure in the flue gas is equal to the CO_2 partial pressure in equilibrium with the CO_2 loading in the solvent, the gas and the liquid phases are considered to be in equilibrium and no further mass transfer will occur. All other parameters being equal, reaching equilibrium at the bottom of the absorber would result in the minimum thermal energy input for solvent regeneration, i.e. minimum specific reboiler duty, as CO_2 uptake would be maximised while sensible and latent heat addition to the water content of the solvent mixture is minimised. However, due to the decreased driving force of absorption as the system approaches equilibrium, an infinitely large absorber would be required. In this study, the packed bed height in the CO_2 absorber is designed so that the rich solvent loading approaches 87.5% of the solvent loading at equilibrium with CO_2 partial pressure in the flue gas at the bottom of the absorber (ca. 0.47mol CO_2/mol MEA). The specific reboiler duty is expected to always be reduced at higher rich loadings [14].

For zero residual emission configurations and the lean loading values used in this study, an intercooler is required between the two packed beds of the absorber. The MEA solvent is extracted from the absorber for cooling, thus increasing the capacity of the solvent to absorb CO_2. The solvent passing through the intercooler is cooled down to a pre-set temperature using water before re-admitting it to the absorber. This ensures that sufficiently high rich loadings are achieved. The rich solvent leaving the bottom of the absorber is pumped to the CO_2 desorber. Prior to the desorber, sensible heat from the hot lean solvent leaving the desorber at ca. 120-130°C is transferred to the rich solvent in the cross-flow heat exchanger. Heat is transferred to the reboiler by condensing low pressure steam extracted from the power cycle associated with the SMR. The heat input required is referred to as the reboiler duty and is usually reported in specific terms, as defined by Equation 4 [23].

$$q_{reg} = \frac{\dot{Q}_{reboiler}}{\dot{m}_{CO2}} = q_{sen} + q_{vap,H2O} + q_{abs,CO2} \qquad (4)$$

Where q_{reg} is the specific reboiler duty for solvent regeneration, $\dot{Q}_{reboiler}$ is the total thermal input to the reboiler, $\dot{m}_{CO2}$ is the mass of CO_2 captured, q_{sen} is the sensible energy necessary to heat the solvent, $q_{vap,H2O}$ is the energy associated with evaporated water leaving with the CO_2, and $q_{abs,CO2}$ is the thermal energy associated with CO_2 desorption from the solvent. The contribution of these three components to the specific reboiler duty varies with rich and lean solvent loadings and, by extension, with the CO_2 capture fraction. Sensible energy is the thermal energy input required to raise the rich solvent temperature from the outlet of the cross-flow heat exchanger to the desorber operating temperature and is primarily a function of heat exchanger pinch temperatures and solvent flow rates (noting that the lean solvent mass flow rate is inherently lower than the rich solvent mas flow rate). The energy contribution to water evaporation is a function of the desorber pressure and the CO_2 exit temperature at the top of the desorber column plus the water/MEA/CO_2 vapour-liquid equilibrium (VLE), which then determines the amount of unrecoverable latent heat of vaporisation of H_2O, which then has to be removed during subsequent condensation and H_2O removal from the CO_2 stream. The third component is the CO_2 desorption energy requirement, which depends on the specific enthalpy of absorption/desorption of the solvent.

After leaving the desorber, the lean solvent enters the solvent heat exchanger for cooling prior to entering the top of the absorber while the CO_2/H_2O mixture leaves the top of the desorber and the water is condensed to achieve a CO_2 % vol of ca. 95%. The CO_2 rich stream is compressed using a three-stage compressor with intercooling (where most of the remaining water is removed) to above the critical pressure of 73.8 bar, and then it is pumped up to the required pressure (assumed to be 110 bar in this study) for dense phase transport and storage, with a CO_2 purity of ca. 99.9%. No advanced heat recovery techniques to reduce the energy penalty are included in this model.

When the CO_2 capture fraction increases the total quantity of CO_2 absorbed by the solvent per second increases. Ultra-high capture fractions in excess of 99% can be achieved by two means, which can be combined: increasing the solvent flow rate, and by extension q_{sen}, or by increasing the solvent capacity, i.e. the difference between lean and rich solvent loadings (which, in practice, means reducing the lean loading by reducing the partial pressure of CO_2 at the base of the desorber). An increase in the absorber packing height relative to that for a lower CO_2 capture fraction may also be necessary to achieve sufficiently high rich loadings and minimise specific reboiler duty at ultra-high capture fractions. Additionally, and depending also on the lean loading used, the location along the absorber and absolute quantity of

the temperature bulge, as result of the heat released by the exothermic absorption reaction, may change at ultra-high capture fractions due to the shift in the CO_2 mass transfer profile. This causes the temperature profile along the absorber to change compared to a 90% capture fraction and, for a given lean loading, mass transfer limiting temperature peaks can become more prominent without intercooling.

In general, increased packing height results in reduced specific reboiler duty for a given CO_2 capture fraction. As the packing height increases, the rate of reduction in specific reboiler duty tends to decrease, leading to diminishing returns as the rich loading approaches equilibrium. To the knowledge of the authors, there is no universally accepted criterion to determine absorber height independent of project specific constraints, as it is trade-off between OPEX and CAPEX burden. In this study, the packing height is sized to return a solvent rich loading of approximately 87.5% of the the equilibrium solvent loading, as previously discussed, as it is believed that this may represent a reasonable trade-off between CAPEX-related absorber packing height and OPEX-related reboiler duty.

The redistribution of liquid in an absorber typically takes place every 20 theoretical stages, corresponding to 8m of Mellapak 252.Y packing, to ensure uniform solvent distribution, with 25 theoretical stages, or 10m of Mellapak 252.Y packing, serving as a realistic upper limit (Ausner, Iija "Maximum packed bed height Mellapak 252.Y" (pers. comm., February 28, 2022)). As a result, a limit of 10m of packing per absorber bed is used in this analysis, although in practice this limit varies according to the packing used and must be properly accounted for during the design phase. In order to reduce CAPEX at higher CO_2 capture fractions, a limit of two absorber beds, or a total of 20m packing, is set as a maximum design constraint. This is supported by evidence from the process models which predict a marginal reduction in reboiler duty for increases in packing height above 20m. For 90% and 95% CO_2 capture, absorber packing heights of 12 and 14m respectively are used, again with a rich loading of approximately 87.5% of the equilibrium value. A FEED study completed by Bechtel, for a CO_2 capture retrofit to a coal fired power plant for 90% CO_2 capture with a flue gas concentration of 11.2% vol, considers two packed beds of 7.5m each [24]. Another FEED study, completed by Nexant, for a pilot scale CO_2 capture plant for installation on a natural gas combined cycle with a flue gas concentration of 3.8% vol for 85% CO_2 capture, assumes a packing height of 22m [25]. As required packing height decreases with increasing flue gas concentration for a given CO_2 capture fraction, 12 and 14m of packing for an SMR PCC application with 20% vol CO_2 in the flue gas appears to be an acceptable estimate.

Solvent return temperature to the absorber is tightly controlled to maximise the CO_2 absorption along the length of the absorber column while maintaining the water balance in the absorber to reduce water uptake or loss from the solvent. As MEA degradation rates increase with temperature, a maximum desorber temperature of 125°C is used in all cases. This is currently considered appropriate for MEA [14, 26-28]. According the model predictions, this maximum temperature allows a lean loading of 0.16 mol CO_2/mol MEA to be achieved at a pressure of 210 KPa without increasing specific reboiler duty due to excessive water vapour in the CO_2 exiting the desorber. Higher desorber operating pressures, and hence temperatures, would facilitate achieving lower lean loadings, which may lead to a more economical CO_2 capture process; further research is, however, required to quantify MEA degradation rates at temperatures above 125°C and also as a function of lean loading and other plant-specific parameters. For example, Braakhuis et al. (2022) recently published MEA thermal degradation data suggesting that reducing lean loading may partially offset the increase in thermal degradation associated with higher operational temperatures [29].

2.2   Levelised Cost of Electricity & CO₂ Capture

The Levelised Cost of X is the discounted lifetime cost of building and operating a production asset, where X is the product, i.e. hydrogen or CO_2. It covers all costs to the producer including, CAPEX, OPEX, fuel, waste disposal and financing costs. The LCOX is expressed as a net present cost (as shown by Equation 2).

$$LCOX = \frac{NPV\ Costs}{NPVX} \qquad (2)$$

$$NPV\ Costs = \sum_n \frac{CAPEX_n + OPEX_n}{(1 + Discount\ Rate)^n} \qquad (3)$$

$$NPV\ X = \sum_n \frac{X\ Production_n}{(1 + Discount\ Rate)^n}$$
$$(n = time\ period)$$

(4)

Where NPV is the net present value of the expense or revenue in question.

A detailed multi-level factorial cost model, as shown in Equation 7, is presented and used to calculate the required CAPEX with an estimated accuracy of +35%/-15% (AACE Class 4) [30] and presented in Table *1*. Engineering, Procurement and Construction (EPC) cost for an CCGT is estimated from a report issued by the BEIS [31]. Total CCS EPC cost is estimated on an equipment level using Equations 6 and 7. Reference costs and scaling parameters for each post combustion CO_2 capture plant item along with installation and tax & freight factors are taken from Sinnott and Towler, R. Woods [32] and the available FEED studies [24, 26, 27] and converted to UK 2020 prices using the chemical engineering plant index (CEPI) [33], historical currency exchange rates and the international construction cost index (ICCI) [34]. A detailed list of economic parameters is available in Appendix A.

$$SC = RC * \left(\frac{SP}{RP}\right)^{Exp}$$

(5)

$$EPC = (f_{contractor\ fee} + f_{contingency}) * \left\{ \begin{array}{c} f_{project\ management} * \\ (f_{civil} + f_{utility} + f_{electrical}) * \\ \left[\sum \left(I + \left(f_{installation} + f_{\frac{tax}{freight}} \right) * SC \right) \right] \end{array} \right\}$$

(6)

$$CAPEX = (EPC_{SMR} + EPC_{PCC}) * \\ \left(\begin{array}{c} 1 + f_{owners} + f_{regulatory} + f_{spares\ \&\ start-up} + \\ f_{connections} + f_{consultancy} \end{array} \right) \\ + Intrest\ during\ construction$$

(7)

Where Exp is the Scaling Exponent, RC is the reference plant cost, RP is the reference plant parameter, SC is the scaled plant cost, SP is the scaled plant parameter, f_i represents a factorial cost and I is the equipment instrumentation cost.

Table 1 Capital & Operational cost estimates

Item	0% residual emissions (M£)	5% residual emission (M£)	10% residual emission (M£)
Total CAPEX	**1051**	**987**	**969**
PCC EPC	430	379	365
SMR EPC	413	413	413
Interest during construction	72	68	67
Additional CAPEX	136	127	124
Annual OPEX	**485**	**468**	**459**
Fixed OPEX	42	40	39
Variable OPEX	443	428	420

3   Results and discussion

Table *2* provides a summary of the design and operating parameters of the PCC plant for a 5%, 10% and zero residual emissions configuration. Packing heights of 12, 14 and 20 metres are used to illustrate the trade-off

between increased CAPEX and diminishing returns in decreased reboiler duty for the 5%, 10% and zero residual emission configurations, respectively. A lean loading of 0.16 mol CO_2/mol MEA is chosen for all cases.

The lean solvent temperature entering the top of the absorber and the intercooling return temperature have a larger effect on the CO_2 absorption rate throughout the absorber column for higher CO_2 capture fractions and thus these temperatures have been optimised for the zero residual emissions case. Solvent return temperature optimisation and intercooling are found to have a marginal effect for both the 5 and 10% residual emission cases and as such are not included. As similar rich and lean loadings are achieved in all cases, specific reboiler duty is also similar. Due to the increase in the absolute quantity of heat input to the reboiler at higher capture fractions, as the total amount of CO_2 captured per unit of H_2 produced increases, the quantity of ambient air and combustion fuel needed increase proportionately in order to supply sufficient steam for solvent regeneration. This leads to an increased flue gas flow rate and a decreased flue gas CO_2 % vol (primarily due to the increased nitrogen content of the flue gas per unit of hydrogen exported with the additional intake of ambient air) detailed in   Table *2*.

Table 2 Design and operating parameters of the CO₂ capture plant

Parameter	Unit	0% residual emissions	5% residual emission	10% residual emission
Flue Gas				
Flow Rate	*kg/s*	275	262	256
Inlet Temperature	*°C*	40	40	40
CO_2 Concentration	*Mole Frac*	19.4	19.9	20.2
Absorber				
Absorbers	-	1	1	1
Packing Stages	-	2	2	2
Packing Height	*m*	20	14	12
Diameter	*m*	12	12	12
Packing Volume	*m³*	2262	1583	1357
Solvent Return Temp	*°C*	38	35	35
Intercooler Return Temp	*°C*	25	-	-
Absorber Flooding	*%*	78	79	79
Rich/Lean HX approach temperature	*°C*	10	10	10
Desorber				
Lean Loading	*mol CO₂/mol MEA*	0.16	0.16	0.16
Rich Loading	*mol CO₂/mol MEA*	0.466	0.469	0.469
Operating Pressure	*KPa*	210	210	210
Reboiler Temperature	*°C*	125	125	125
Specific Reboiler Duty	*GJ/tCO₂*	3.67	3.62	3.60

Table 3 shows the performance of the SMR operating with zero residual emissions compared to 5% and 10%. At zero residual emissions, the additional steam for solvent regeneration requires an increase in the supplementary fuel flow rate. This results in a marginal shift in the H_2 to Power production ratio. A 1.9% point difference in H_2 production efficiency (HHV) is noted between the 5% residual and zero residual emissions cases. House load increases with increases CO_2 capture fraction, primarily due to the higher CO_2 flow rate to the compressors. No other process modifications are required to reach this increased CO_2 capture fraction.

Table 3 Performance assessment of a SMR plant with PCC at zero, 5% and 10% residual emission.

Parameter	Unit	0% residual emissions	5% residual emission	10% residual emission
H_2 Export (HHV)	MW_{th}	1000	1000	1000
Net Power Output	MW_e	33	30	28
House Load	MW_e	26	24	23
Total Fuel (HHV)	MWh_{th}/s	0.416	0.404	0.400
H_2 Production Eff (HHV)	%	66.8	68.7	69.6
CO_2 Export	kg/s	76	71	67
Direct CO_2 emissions	gCO_2e/MJ LHV	0.0	4.3	8.6

An economic analysis predicting the levelised cost of hydrogen (LCOH) and the levelised cost of capture (LCOC) is shown for zero, 5% and 10% residual emissions operation of a 1GW capacity SMR when no carbon pricing is applied, the results are detailed in _Table 4_. Under these conditions zero residual emission operation is shown to be just 3.2£/MWh$_{th}$ HHV and 2.2£/MWh$_{th}$ HHV more costly than hydrogen produced with 10% and 5% residual emissions respectively. However, a zero-price for emitted CO_2 is unlikely to be consistent with climate mitigation targets and these cases are presented for illustrative purposes only. A carbon price of 172£/tCO$_2$ is found to be the point at which H_2 production with zero residual emissions and H_2 production with 5% residual emissions are equal in LCOH. _Table 4_ shows that the LCOH and LCOC follows a non-linear relationship to capture fraction; this is tentatively attributed to non-linear CAPEX relationships, increase fuel requirements and varying specific reboiler duty.

Table 4 Levelised cost of hydrogen and CO_2 captured for Zero, 5% and 10% residual emission operation

		0% residual emissions	5% residual emission	10% residual emission
LCOH	**£/MWh$_{th}$**	**62.0**	**59.8**	**58.8**
Fuel	£/MWh$_{th}$	41.9	40.8	40.3
CAPEX	£/MWh$_{th}$	12.7	12.0	11.7
OPEX	£/MWh$_{th}$	5.1	4.8	4.7
Emissions	£/MWh$_{th}$	0.0	0.0	0.0
CO_2 Transport and storage	£/MWh$_{th}$	5.3	4.9	4.6
Power Generation Revenue	£/MWh$_{th}$	-3.0	-2.7	-2.5
LCOC	**£/tCO$_2$**	**54.1**	**52.4**	**53.2**

4   Conclusions and future work

Achieving climate compatible hydrogen produced from natural gas will require industry and blue hydrogen project developers to design plants that are capable of capturing 100% of fossil CO_2 emissions. We combine insights from process modelling with cost data from publicly available Front End Engineering Design studies to develop a techno-economic model of a steam methane reformer (SMR) equipped with post-combustion CO_2 capture. It is estimated that capital and operating costs increase by 6.5% and 3.7%, respectively, for a 1GW HHV zero residual CO_2 emission hydrogen facility, compared to hydrogen produced with 5% residual CO_2 emissions. The Levelised Cost of Hydrogen with zero residual emissions is 62£/MWh$_{th}$ HHV for 2020 UK capital costs and a natural gas price of 28 £/MWh$_{th}$ HHV.

The PCC plant designed to achieve zero residual emissions requires a higher structured packing section in the CO_2 absorber. It increases from 14 m, for 5% residual emissions configuration, to 20 m of structured packing in order to limit the specific reboiler duty to a minimal increase of 1.6% on a per tonne of CO_2 basis. The additional gas input needed to increase in CO_2 captured per unit of H_2 produced lowers hydrogen production efficiency by 1.9% point HHV, from 68.7 %, for 5% residual emissions configuration, to 66.8%, for zero residual emissions configuration. This work builds on recent studies, referenced in this article, to provide compelling evidence that, when the CO_2 absorber

design is optimised for ultra-high CO_2 capture fractions, the increase in cost and energy penalty associated to the CO_2 capture process in steam methane reforming hydrogen production is significantly smaller than previously thought.

However, ensuring zero residual CO_2 emissions at the point of production is only the first step to achieving net zero hydrogen production from natural gas. Future work must first quantify and then identify pathways to reduce to zero or permanently offset with negative emission technologies any remaining greenhouse gas emissions in the supply chain.

Acknowledgements

Appendix A. Economic Model Parameters

Parameter	Unit	Value
Discount rate [35]	%	7.8
Interest Rate [26]	%	4.0
Productive lifetime [19]	Years	25
Base year	-	2020
Year operational	-	2023
Build Time [19]	Years	3 Years
CAPEX expenditure curve [19]	%	20/45/35
Load Factor [19]	%	95%
Transport and storage cost [31]	£/tCO$_2$	19
Natural Gas Fuel price [35]	£/MWh	28
Electricity selling price [36]	£/MWh	90
Chemical Engineering Plant Index (CEPCI) [33]	-	596.2
International Construction Cost Index [Netherlands/UK/Texas] [34]	-	100/136/86
USD to GBP exchange rate (2020) [37]	£/$	0.78
Equipment Tax/Insurance/Freight [26, 32]	%	10
Civil Works factorial cost [24, 26, 27]	%	21
Utilities factorial cost [24, 26, 27]	%	18
Electrical factorial cost [24, 26, 27]	%	30
Project management [24, 26, 27]	%	21
Contractor Fee [32]	%	3
Project Contingency [24, 26]	%	10
Owners Costs [35]	%	7
Start-up & Spares [35]	%	5
Utility Connections [35]	%	1
Consulting [35]	%	1
Maintenance (CCS) [31]	%/Year	1.5
Maintenance (SMR) [31]	%/Year	3.0
Labour [31]	£/Employee/Year	69,810
Insurance/Tax/Admin [35]	%/Year	1.5
Regulatory [35]	%	2.0
MEA [14]	£/t	940
Caustic [38]	£/t	34
Reclaimer disposal [14]	£/t	500
Working Capital [31]	-	1 Months Consumables

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