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

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ABSTRACT

This article challenges the view that zero carbon hydrogen from steam methane reforming (SMR) is prohibitively expensive and that the cost of CO_2 capture increases exponentially as residual emissions approach zero; a flawed narrative often eliminating SMR produced hydrogen as a route to net zero. We show that the capture and geological storage of 100% of the fossil CO_2 produced in a SMR is achievable with commercially available post-combustion capture technology and an open art solvent. The Levelised Cost of Hydrogen (LCOH) of 69£/MWh_{th} HHV (2.7£/kg) for UK production remains competitive to other forms of low carbon hydrogen, but retains a hydrogen lifecycle carbon intensity of 5 gCO_2e/MJ (LHV) due to natural gas supply chain and embodied greenhouse gas (GHG) emissions. Compensating for the remaining lifecycle GHG emissions via Direct Air Capture with geological CO_2 Storage (DACCS) increases the LCOH to 71–86 £/MWh_{th} HHV (+3–25%) for a cost estimate of 100–1000 £/tCO_2 for DACCS and the 2022 UK natural gas supply chain methane emission rates. Finally, we put in perspective the cost of CO_2 avoidance of fuel switching from natural gas to hydrogen with long term price estimates for natural gas use and DACCS, and hydrogen produced from electrolysis.

1. Introduction

Hydrogen is increasingly being recognised globally as a useful component of a net zero greenhouse gas (GHG) economy, since no CO_2 is produced at the point of use. It is a potential energy vector for decarbonising decentralised emission sources such as home heating and heavy goods vehicles (HGV), or industries where electrification or direct CO_2 capture is impractical (Joffe et al., 2018).

The need for rapid scale up of low GHG emission hydrogen production has been reflected in the Sixth Carbon Budget (Stark et al., 2020), issued by the UK Committee on Climate Change, and the 2021 UK hydrogen strategy (UK Hydrogen, 2021). This report 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 (Heap et al., 2016), 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 (Beis 2022) 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 direct CO2 intensity of 0.1 gCO_2e (equivalent)/MJ H2 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 (UK Hydrogen, 2021; Hydrogen production costs 2021).
- Methane reforming with CO_2 capture and storage (blue hydrogen): This includes a Carbon Capture & Storage (CCS) system, which separates the CO_2 produced during the process for permanent geological storage. This process may be done in a Steam Methane Reformer (SMR) or an Autothermal Reformer (ATR). The carbon intensity of the hydrogen produced varies depending on the CO_2 capture fraction applied. Typical reported values lie between 0 – 21.4 gCO_2e/MJ H2 (LHV) (UK Hydrogen, 2021). In the past, lower capture fractions (~90%) have been assumed for SMR+CCS and higher values (~95%) for ATR+CCS (e.g. see (Hydrogen production costs

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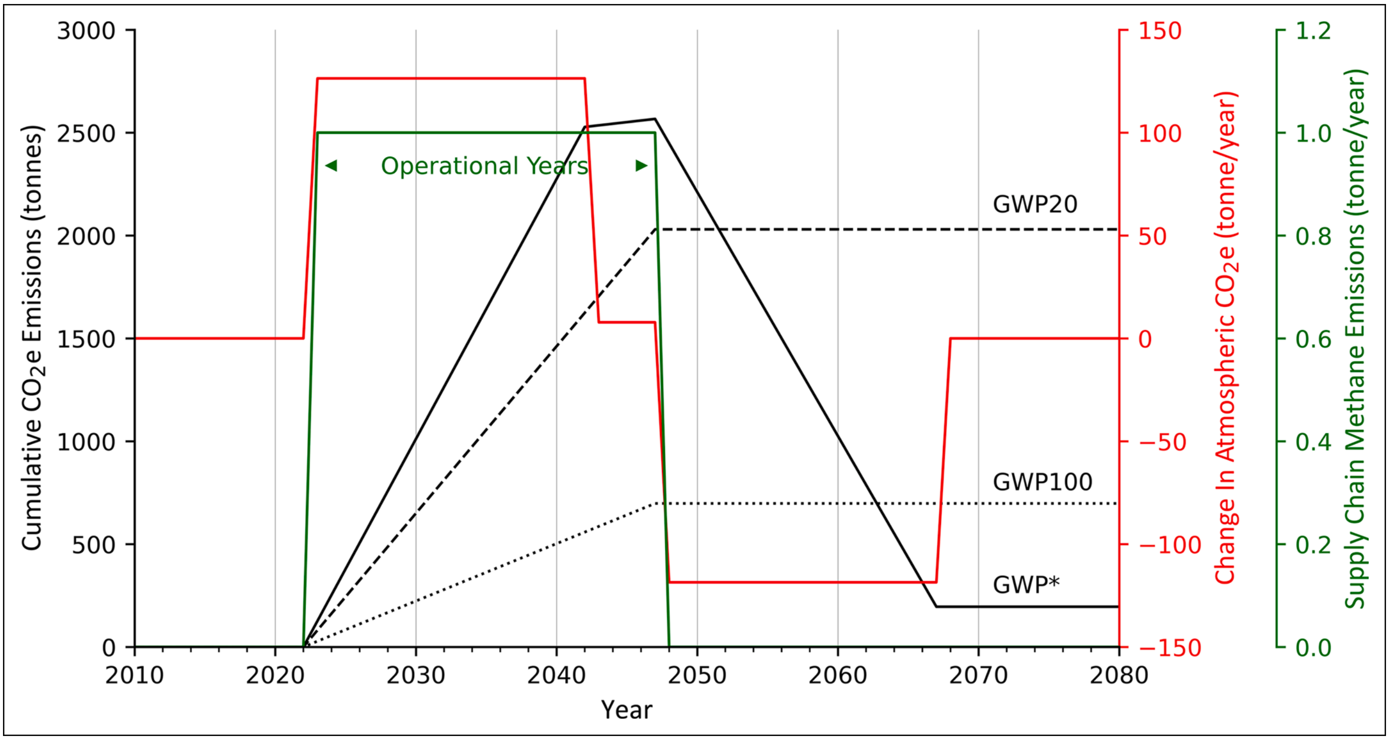


Fig. 1. Comparison of GWP*, GWP100 & GWP20 when evaluating cumulative CO_2e emissions resulting from a sustained methane emission rate of 1 tonne per annum over a 25 year period. The green line represents annual methane emission's while the red line shows net change in atmospheric CO2e per year as predicated by GWP*.

2021)). 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 direct CO2 intensity of 8.7 gCO_2e/MJ H_2 (LHV)(Hydrogen production costs 2021).

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 SMRs is often assumed to be 90% and, with the assumed 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 blue hydrogen as a climate mitigation technology in the context of an energy system compatible with net zero GHG ambitions.

Indeed this is the case in a recent article published by Howarth and Jacobson (Howarth and Jacobson, 2021) claiming that uncaptured CO_2 emissions from the SMR process, coupled with the energy required to drive the CO_2 capture process and the upstream methane emission result in a carbon intensity of blue hydrogen of 88–91% that of an SMR with no CO_2 capture. The assumptions made by Howarth and Jacobson are widely criticized by Romano et al. (Romano et al., 2022). As an illustration, the SMR model proposed by Howarth and Jacobson assumes a net CO_2 capture fraction of just 61%, an average capture fraction of 76% is applied to the SMR with no allowance for the capture of the CO_2 generated due to the additional energy requirement imparted by CO_2 capture, a critical oversight. Despite the low CO_2 capture fraction, a thermal energy consumption rate in the CO_2 capture plant of 6.25 GJ/tCO_2 captured is implicit in the analysis. This is assumed to be supplied via the combustion of additional methane with no thermal integration to the SMR plant or supplementary electricity generation, as such, the thermal energy used is high grade and a poor application of the 2nd law of thermodynamics. This represents a dramatic deviation from the low grade thermal energy, typically low pressure steam, requirement reported in operational plants. Nessi et al. (Nessi; et al., 2021) recently completed a review of 37 PCC plants in or previously in operation worldwide. 25 plants had sufficient data available in the public domain and reported an achieved specific thermal energy requirement for CO_2 capture of between 2.1–3.7GJ/tCO_2, with CO_2 capture fractions ranging from 45% in one case to up to 98%, with circa 90% being typical.

This article extends on Romano et al. (2022) to add evidence on the integration, design, operation and costs of hydrogen production with 100% CO_2 capture, and the added costs of permanent offsetting to achieve zero lifecycle carbon intensity. It is timely with a growing body of research on ultra-high CO_2 capture operation, and timely with regulatory developments pushing for increasing capture fractions in the permitting of new CCS facilities.

Recent UK guidance by the UK Environment Agency (Gibbins and Lucquiaud, 2021; Gibbins and Lucquiaud, 2021) notably 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 (Curtis, 2022). 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 (Comparison of commercial 2022). 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 (UK 2022). As this standard proposes the functional unit of CO_2 intensity of hydrogen to be gCO_2e/MJ H_2 (LHV) it will be reported as such in this study for comparative purposes.

The 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 is reported in Feron et al. (2019); Danaci et al. (2021); Michailos and Gibbins (2022); Gao et al. (2019); Ismail Shah et al. (2021); Hirata et al. (2019). Feron et al.

reported that increasing the CO_2 capture fraction of a post-combustion solvent-based system (30%wt MEA) from 90% to a zero residual emission level would give a 1.5 percentage point reduction (34.5% to 33%) in thermal efficiency on a 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) (Feron et al., 2019). Work completed by Danaci et al. reported that, for a representative flue gas flow rate of 500 kg/s, a 30%wt MEA solvent and flue gas CO_2%vol concentrations of 4%, 10% and 20%, transitioning from 90% to 99% CO_2 capture results in an increase in total capture cost of 7%, 10% and 13% respectively (Danaci et al., 2021). 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 a 35%wt solvent solution, providing the absorber is sufficiently sized and the desorber pressure is high enough to avoid excessive water vapour in the product CO_2 at the reduced lean loading required (Michailos and Gibbins, 2022). 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 (Gao et al., 2019). Tests completed at Technology Centre Mongstad show that, with 24 m 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 (Ismail Shah et al., 2021), 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) (Hirata et al., 2019).

When considering the life cycle emissions of hydrogen production from natural gas, it is also essential to account for all aspects of the hydrogen supply chain; venting, flaring and pipeline natural gas emissions are potentially large contributors to the carbon intensity of blue hydrogen.

While methane is a more potent greenhouse gas than CO_2, it is shorted lived in the atmosphere with an estimated perturbation residence time of 11.8 ± 1.8 years (Masson-Delmotte et al., 2021) and, as such, does not accumulate in the in the same way. If, for example, methane emissions from the previous decade were equal to current emissions virtually no additional contribution to the rate of warming would have occurred because the level of atmospheric methane remains approximately constant. Research completed by Oxford University (Smith et al., 2021; Cain et al., 2019) suggests that the use of Global Warming Potential (GWP) values consistently overestimates the warming effect of methane in scenarios where global methane emissions are constant or reducing and recommends the use of an alternative metric called GWP*. This metric relates current or forecasted annual methane emissions to historical values and a CO_2e value for the net change in atmospheric methane is predicted. CO_2e values under GWP* are calculated as shown in Eq. (1). Climate modelling (Smith et al., 2021; Cain et al., 2019) studies suggest that GWP* correlates with global temperature response to methane emissions to a much higher degree of accuracy than GWP.

$$CO_2e(t) = GWP_{100} * [4.53 * E_{CH_4}(t) - \ 4.25 * E_{CH_4}(t-20)] \tag{1}$$

Where t is the year under analysis, $CO_2e(t)$ is the equivalent CO_2 emissions resulting from methane emissions in year t, GWP_{100} is the global warming potential value of methane over 100 years and $E_{CH4}(t)$ is the total annual methane emissions at year t.

Fig. 1 shows the predicted cumulative CO_2e emissions resulting from a sustained 1 tonne of annual methane emissions over a 25 year period when calculated using GWP*, GWP100 and GWP20. Fig. 1 illustrates that the GWP100 and GWP20 metrics fail to capture the short lived nature of methane emissions and assume that the resulting CO_2e emissions remain present in the atmosphere indefinitely. GWP*, however, shows a spike in CO_2e emissions in excess of that predicted by GWP100 & 20 in the first 20 years of operation. The rate of emissions then drops off in the final 5 years of operation and, as the historical methane emissions oxidise. A rapid decline in cumulative atmospheric CO_2e is seen once methane emissions cease, eventually returning to a net change in atmospheric CO_2e of 195tCO_2e, much lower than the 678 & 2030 tCO_2e predicted by GWP100 & 20 respectively. For simplicity GWP* assumes all oxidation of atmospheric CH_4 occurs at year 20, in reality there is an exponential decrease in of the quantity of atmospheric CH_4 over time, the end result being a reduction in the peak atmospheric CO_2e presented in Fig. 1. Fig. 1 illustrates how GWP100 and GWP20 (as used by Howarth and Jacobson (Howarth and Jacobson, 2021)) can overestimate the long term net climate impact of methane emissions, hence GWP*, as a more accurate metric, is used in this work.

It is worth noting that, although the net cumulative CO_2e predicted by GWP* eventually falls to 195tCO_2e per tonne of sustained methane emissions over 25 years, temporary driving of global warming in excess of what could be attributed to this long term value will occur in the period of elevated atmospheric CO_2e i.e. during and immediately after cessation of methane emissions. This temporary warming rate increase requires consideration during the environmental impact assessment of a proposed project and highlights the importance of efforts to reduce fugitive methane emissions from supply chains.

A recent review on the climate impacts of blue hydrogen by Bauer et al. (Bauer et al., 2022) characterises global natural gas supply chain emission rates as “grams of CH_4 emitted per gram of natural gas delivered”. Large variability is noted on both spatial and temporal bases, with the emission rate and measurement accuracy also varying considerably based on the producer in question. Near zero emissions of 0.1% is reported in regions such as Norway and Qatar while emission factors of over 6% are reported in areas such as Libya and Iraq. Bauer et al. used a factor of 1.5% for a mid-range value in their life cycle assessments. Additionally, the recent low carbon standard issued by the UK BEIS advises using an average carbon intensity of natural gas supply of 6.29 gCO_2e/MJ (LHV) (calculated using GWP100) for the UK natural gas supply chain (UK 2022), which corresponds to an emissions rate of 1.04%. In a net zero economy, where any remaining life cycle emissions will have to be compensated for via the use of Carbon Dioxide Removal Technologies (CDRs), as the likely lowest cost option these supply chain emissions will need to be reduced as far as reasonably practicable, a goal echoed by the recent COP26 pledge to reduce global methane emissions by 30% by 2030 (Cop26, 2021).

An important concept in CDRs is the permanence of CO_2 storage away from the atmosphere. Climate change is likely to be a millennium spanning challenge due to the very long lifetime of CO_2 in the atmosphere and the inertia of the climate response to rising atmospheric CO_2 concentrations, and thus any negative emission technology must be consistent with a long term mitigation strategy. Nature-based negative emissions via e.g. reforestation, soil management have uncertain permanence compared to the use of engineered technologies such as Bioenergy with CCS (BECCS) and Direct Air Capture (DACCS) that place the CO_2 in secure geological storage with expected retention of over 10,000 years (Alcalde et al., 2018). For the purposes of this paper DACCS is used as the CDR technology for comparisons, since it is less dependant on finite ecosystem limitations than BECCS. Since DACCS has a higher reported cost than most other CDR technologies (Fuss et al., 2018), this also sets a cap on the premium for permanently compensating for indirect emissions in the hydrogen production chain. Due to limited commercial deployment to date, evidence-based cost estimates for future DACCS are currently unavailable. A recent review by McQueen et al. investigates the current state of the art in DACCS technology (Mcqueen et al., 2021). They place the near-term cost of commercial DACCS (excluding transport and geological storage costs) at

350–400£/tCO_2 and conclude that prices are expected to fall as DACCS deployment increases and the “learn by doing” effect is realised. A cost range of 100–1000£/tCO_2 with a median value of 300£/tCO_2 is used in this paper; it is considered to cover a band consistent with values reported by the International Energy Agency (Direct Air Capture 2022).

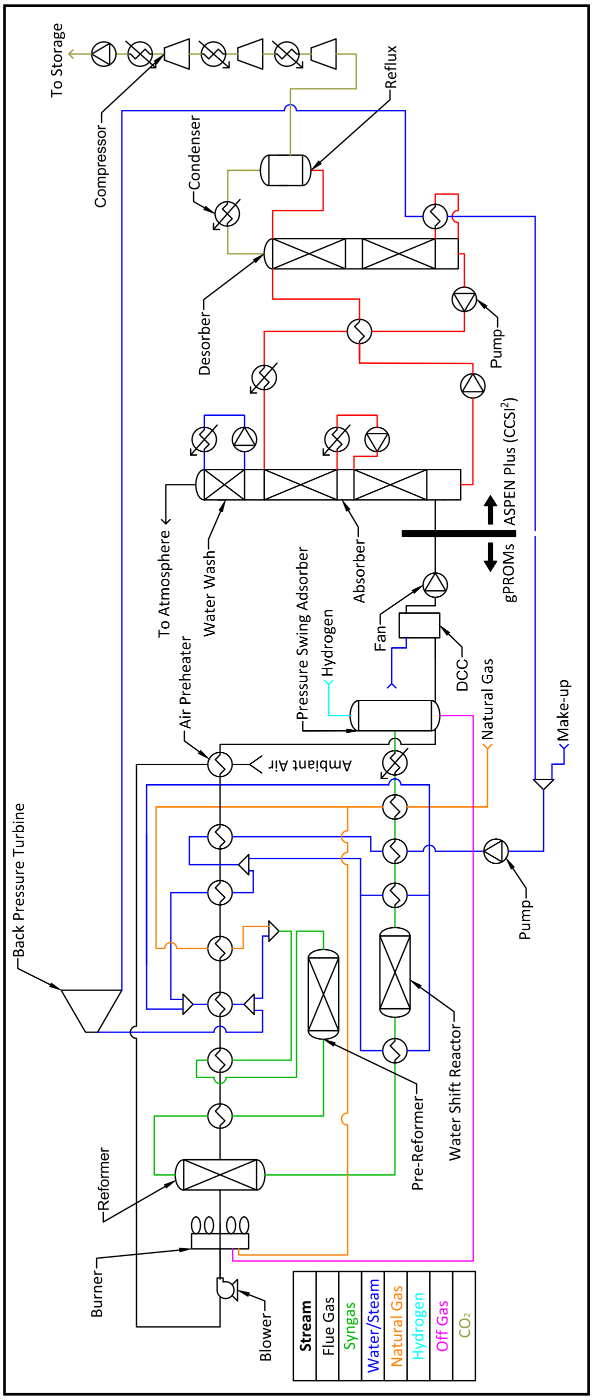


Fig. 2. Steam methane reformer with post combustion carbon capture configuration.

The techno-economic analysis of a conventional steam methane reformer (SMR) in this paper includes three post-combustion capture (PCC) plant options that are designed and optimised for distinct CO_2 capture fractions:

- Net 90% CO_2 capture with 10% residual emissions, corresponding to a production carbon intensity of 8.6 $gCO_2e/MJ\ H_2$ (LHV). It is included to provide comparable evidence with previous studies.
- Net 95% CO_2 capture with 5% residual emissions, corresponding to a production carbon intensity of 4.3 $gCO_2e/MJ\ H_2$ (LHV). This is included to keep in line with recent regulatory developments in the UK for the permitting of post-combustion capture units.
- Zero residual emissions, i.e. 100% capture of the fossil carbon entering the boundaries of the hydrogen plant, with a production carbon intensity of 0.0 $gCO_2e/MJ\ H_2$ (LHV). It corresponds to an overall CO_2 capture fraction of 99.8% in the CO_2 capture plant, once atmospheric CO_2 entering the boundaries of the plant in the combustion air is accounted for.

A design of a PCC plant for ultra-high capture fractions from SMR flue gas is presented for the first time, drawing on state of the art research (Feron et al., 2019; Danaci et al., 2021; Michailos and Gibbins, 2022; Gao et al., 2019; Ismail Shah et al., 2021; Hirata et al., 2019), front end engineering design (FEED) studies (Elliott, 2022; Bechtel 2019; Nexant 2016; Bechtel 2018) and computational modelling (Morgan et al., 2020). Results from this PCC model are coupled with a SMR model to establish the additional thermodynamic penalty associated with transitioning to zero residual emission operation.

The output of a detailed multi-level factorial cost model used to predict plant capital expenditure (CAPEX) and operational expenditure (OPEX) feeds a comprehensive economic analysis estimating the Levelised Cost of Hydrogen (LCOH), Levelised Cost of Capture (LCOC) and Cost of CO_2 Avoided (CCA). The cost of producing zero carbon hydrogen on a life cycle basis, with DACCS with geological storage compensating for natural gas supply chain emissions, embodied carbon in construction and CO_2 transport and storage emissions is presented. A sensitivity study establishes to what degree variation in key economic and operational factors effects the LCOH, providing a clear pathway for further cost reductions and de-risking of investments.

2. Methodology

2.1. Modelling methodology for a steam methane reformer (SMR) with post combustion CO_2 carbon capture (PCC)

A model of a 1000 MWh_{th} HHV SMR is produced in gPROMS Process Builder (gPROMS products 2021), a process modelling platform that allows the creation of bespoke unit models for each specific plant operation, Peng-Robinson & IAPWS-95 are used as the gas mixture and steam/water thermodynamic models respectively. The flue gas and low pressure steam conditions is feed into an open source steady state MEA based PCC model (Morgan et al., 2020). This model (Morgan et al., 2020) was created by the Carbon Capture Simulation for Industry Impact ($CCSI^2$) partnership (Ccsi 2022) in Aspen Plus (Aspentech 2022), which was developed using pilot scale data provided by the US National Carbon Capture Centre (NCCC) (National 2021). The output from the PCC model, namely thermal and electrical energy requirements, is then feed back into the SMR model and an iterative design optimisation process is completed.

The SMR with PCC process is illustrated in Fig. 2, the technical and operational parameters are based on a conventional SMR with post-combustion CO_2 capture in a report commissioned by the IEA Greenhouse Gas R&D Programme (IEAGHG) in 2017 (Collodi et al., 2017). A detailed list of plant parameters are available in Appendix A.

2.2. Flue gas path

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 pressure swing adsorber 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 being sent to the post-combustion CO_2 capture plant for CO_2 removal.

The flue gas exits the air preheater in Fig. 2 and initially passes through a direct contact cooler, 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 then 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. The now CO_2 rich solvent (rich solvent) leaves the bottom of the absorber to be stripped of CO_2.

If we were to assume constant CO_2 partial pressure in a flue gas, for illustrative purposes only as this is not possible in actuality, the driving force for absorption would decreases as the solvent absorbs CO_2, resulting in a decreased mass transfer rate. When the CO_2 partial pressure in the flue gas is equal to the CO_2 equilibrium pressure above the solvent, the flue gas and the solvent are considered in equilibrium and no further mass transfer will occur. All other things being equal, 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. Specific reboiler duties are expected to always be reduced at higher rich loadings (Michailos and Gibbins, 2022), however, 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.

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 developed as part of this study that predict a marginal reduction in reboiler duty for increases in packing height above 20m.

For capture fractions resulting in zero residual emission configurations, given the lean loading values used in this study, an intercooler is found to be required between the two packed beds of the absorber. This extracts the MEA solvent 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 cooling 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 Eq. (2) (Soltani et al., 2017).

$$q_{reg} = \frac{H_{reboiler}}{\dot{m}_{CO_2}} = q_{sen} + q_{vap,H_2O} + q_{abs,\ CO_2} \tag{2}$$

Where q_{reg} is the specific reboiler duty for solvent regeneration, $H_{reboiler}$ is the total thermal input to the reboiler, $\dot{m}_{CO_2}$ is the mass of CO_2 extracted from the solvent, q_{sen} is the sensible energy necessary to heat the solvent, q_{vap,H_2O} is the energy associated with evaporating water leaving with the CO_2, and $q_{abs,\ CO_2}$ 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 solvent rich and lean 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. It 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 mass 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). This 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.

As MEA degradation rates increase with temperature, a maximum desorber temperature of 125 °C applied to all cases. This is currently considered appropriate for MEA (Michailos and Gibbins, 2022; Elliott, 2022; Bechtel 2019; Léonard et al., 2015). According to the model predictions, this 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 or entirely offset the increase in thermal degradation associated with higher operational temperatures (Braakhuis et al., 2022) as thermal degradation in MEA appears to be rate limited by CO_2 concentration in 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 then

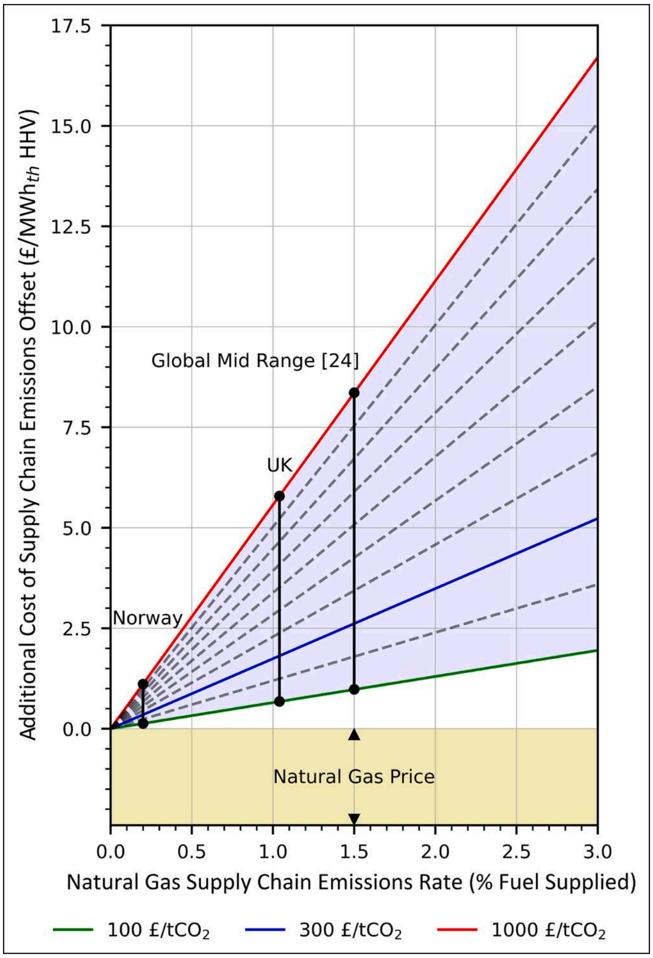


Fig. 3. The additional cost to natural as price to offset supply chain emissions, for a range of supply chain emissions and costs of direct air capture under GWP*.

assumed to be compressed in this study using a three-stage compressor with intercooling (where most of the remaining water is removed) to above the critical pressure of 73.8 bar, where it may be liquefied and pumped up to the required pressure (assumed to be 110 bar in this study) for transport and storage, with a CO_2 purity of ca. 99.9%. No additional heat recovery techniques to reduce the energy penalty are included in this model.

When CO_2 capture fractions are increased, 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 difference between lean and rich solvent loadings, and thus increasing the solvents capacity to absorber CO_2 per kg (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 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 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 can cause 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 in the absence of intercooling.

2.3. Natural gas reforming and power generation

High pressure steam and preheated natural gas feedstock at 400°C and 34 bar are mixed, further heated to 500°C 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 mol H_2O/mol Carbon 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. 91 °C. The Steam-Methane reforming reaction as described by the equilibrium shown in Eq. (3) and the Water-Gas shift reaction as described by Eq. (4). 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'.

$$CH_4 + H_2O \leftrightarrow CO + \ 3H_2 \ \Delta H_r = \ 206\frac{\text{kJ}}{\text{mol}} \tag{3}$$

$$CO + H_2O \leftrightarrow CO_2 + \ H_2 \ \Delta H_r = \ -41\frac{\text{kJ}}{\text{mol}} \tag{4}$$

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, a 7%, 10% and 5% pressure drop is assumed for the pre-reformer, reformer and water gas shift reactor respectively.

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), which is modelled as a "black box" and wherein it is assumed that 90% of the H_2 (Collodi et al., 2017) is separated out at 25 bar for export. The off gas, consisting of the remaining H_2, unreacted CH_4, H_2O, 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, with an assumed isentropic efficiency of 90%, 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.

2.4. Supply chain emissions

As a potent greenhouse gas, any methane emissions in the natural gas supply chain need to be compensated for via some form of negative emission technology in order for hydrogen production to achieve zero life cycle greenhouse gas emissions. The specific carbon intensity of natural gas supply ranges from 1.1 to 16.4 $kgCO_2eq/MWh_{th}$ (HHV) of natural gas for the emission rate range of 0.2 to 3% when calculated using GWP* for a 25 year operational life. For the purpose of this analysis, we assume that the necessary installed capacity of DACCS is

Table 1
Embodied and CCS upstream CO_2 emissions.

Emission source	Unit	Value
Post Combustion CO_2 Capture Plant Operation (Herraiz Palomino et al., 2022)	gCO_2e/ KG CO_2 Captured	19.2
Post Combustion CO_2 Capture Plant Construction (Herraiz Palomino et al., 2022)	gCO_2e/ KG CO_2 Captured	0.85
Steam Methane Reformer Construction (Spath and Mann, 2001)	gCO_2e/ KG H_2 Produced	41

commercially available and independent of the H_2 producer. A cost range of 100–1000£/tCO_2 is considered to cover a range of estimates consistent with values reported in the literature (Mcqueen et al., 2021; Direct Air Capture 2022) while CO_2 transport and storage cost for the resulting additional captured CO_2 are set at 19 £/tCO_2 (Assessing the Cost 2018) and the natural gas price set at 28£/MWh_{th} HHV (Review of next generation 2022).

The cost of offsetting supply chain emissions can be directly allocated to fuel prices in LCOH calculations by attributing an additional cost per unit of fuel supplied. This is shown in Fig. 3 as an additional fuel price premium for a range of emission rates and DACCS costs. A DACCS cost of 300£/tCO_2 and a UK natural gas supply emission rate of 1.04% is used as the median estimate for LCOH analyses, resulting in a 6% (1.8 £/MWh_{th} HHV) increase in fuel costs. Increasing the long term fuel cost assumed in this study of 28£/MW_{th} to 29.8 £/MW_{th} HHV. A DACCS price of 300 £/tCO_2 is consistent with a report by the US National Academy of Science and Engineering (Negative Emissions 2019) and at the upper end of estimates by the International Energy Agency (IEA) for plants built using current technology at 125–335\$/tCO_2 (Direct Air Capture 2022).

A best case scenario is also included in this study, where methane emissions from the supply chain are reduced to be broadly in line with leaders Norway and Qatar (Bauer et al., 2022) and an average emission rate of 0.2% is assumed.

2.5. *Embodied carbon and CCS fugitive emissions*

Estimated values for the embodied carbon emissions associated with the construction of both the SMR and PCC plant, along with downstream emissions associated with CO_2 transport and storage, are detailed in Table 1. A lifecycle assessment (LCA) on natural gas steam reforming completed by the National Renewable Energy Laboratory (NREL) (Spath and Mann, 2001) provides the estimation for embodied CO_2 in the SMR construction, while Herraiz et al. provides a detailed LCA on the capture plant construction and operational chain (Herraiz Palomino et al., 2022).

The embodied CO_2 emissions from construction are distributed over the construction period of the plant while the CO_2 emissions associated with operation of the CO_2 capture plant and infrastructure are considered to occur annually. All embodied CO_2 emissions are recaptured with DACCS and permanent geological storage. It represents a realistic order of magnitude estimation and demonstrates that the effect on the LCOH is of the order of 1.0–8.6£/MWh_{th} HHV for DACCS costs in the range of 100–1000 £/tCO_2. Of note is the high contribution of reclaimer bottoms disposal to the embodied carbon value (circa 6 gCO_2 eq/$kgCO_2$ captured); Herraiz et al. assume that this waste product is disposed of via incineration with no associated CCS. This is unlikely to be consistent with net zero commitments, and as such the embodied CO_2 emission value used here is likely to be a conservative estimate in the long term. Additionally, as the respective industries involved in SMR & PCC construction (steel, concrete, transport etc.) and MEA production decarbonise, as well as improved solvent management procedures develop it can be expected that the total embodied CO_2 value would decrease proportionally leading to a reducing LCOH penalty over time.

Table 2
Economic model parameters.

Parameter	Unit	Value
Discount rate (Review of next generation 2022)	%	7.8
Interest Rate (Elliott, 2022)	%	4.0
Productive lifetime (Collodi et al., 2017)	Years	25
Base year	–	2023
Year operational	–	2026
Build Time (Collodi et al., 2017)	Years	3 Years
CAPEX expenditure curve (Collodi et al., 2017)	%	20/45/35
Load Factor (Collodi et al., 2017)	%	95%
Transport and storage cost (Assessing the Cost 2018)	£/tCO_2	19
Natural Gas Fuel price (Review of next generation 2022)	£/MWh	28
Electricity selling price (Mullen and Lucquiaud, 2022)	£/MWh	105
Chemical Engineering Plant Index (CEPCI) (THE CHEMICAL ENGINEERING 2022)	–	816.3
International Construction Cost Index [Netherlands/UK/Texas] (Arcadis 2020)	–	100/138/97
USD to GBP exchange rate (2020) (Rates, 2023)	£/\$	0.82
Equipment Tax/Insurance/Freight (Elliott, 2022; Donald, 2007)	%	10
Civil Works factorial cost (Elliott, 2022; Bechtel 2019; Bechtel 2018)	%	21
Utilities factorial cost (Elliott, 2022; Bechtel 2019; Bechtel 2018)	%	18
Electrical factorial cost (Elliott, 2022; Bechtel 2019; Bechtel 2018)	%	30
Project management (Elliott, 2022; Bechtel 2019; Bechtel 2018)	%	21
Contractor Fee (Donald, 2007)	%	3
Project Contingency (Elliott, 2022; Bechtel 2018)	%	10
Owners Costs (Review of next generation 2022)	%	7
Start-up & Spares (Review of next generation 2022)	%	5
Utility Connections (Review of next generation 2022)	%	1
Consulting Fee (Review of next generation 2022)	%	1
Maintenance (CCS) (Assessing the Cost 2018)	%/Year	1.5
Maintenance (SMR) (Assessing the Cost 2018)	%/Year	3.0
Labour (Assessing the Cost 2018)	£/Employee/Year	69,810
Insurance/Tax/Admin (Review of next generation 2022)	%/Year	1.5
Regulatory (Review of next generation 2022)	%	2.0
MEA (Michailos and Gibbins, 2022)	£/t	940
Caustic (Evaluating the Costs of Retrofitting 2017)	£/t	34
Reclaimer disposal (Michailos and Gibbins, 2022)	£/t	500
Working Capital (Assessing the Cost 2018)	–	1 Months Consumables

Table 3
Post combustion CO_2 capture plant engineering, procurement & construction (EPC) cost estimates.

Item	Zero residual emissions (M£)	5% residual emission (M£)	10% residual emission (M£)
Absorber	58	47	45
Regeneration	66	59	55
Heat Exchangers	47	44	42
Compression	39	37	37
Miscellaneous	21	22	22
Tax & Freight	23	21	20
Total Plant Cost	256	230	220
Civil	54	48	46
Utility	46	41	40
Electrical	77	69	66
Project Management	91	82	78
Bare Module Cost	523	470	450
Contractor Fee	16	14	14
Contingency	52	47	45
Engineering, procurement & construction	591	531	509

Table 4
Total Plant CAPEX Estimates.

Item	Zero residual emissions (M£)	5% residual emission (M£)	10% residual emission (M£)
PCC EPC	591	531	509
SMR EPC	605	605	605
Connections	12	11	11
Start-up/ Commissioning/ Spares	60	57	56
Regulatory Costs	24	23	22
Owners Costs	84	79	78
Consultation Costs	12	11	11
Interest during construction	103	97	96
Total CAPEX	1489	1415	1387

2.6. *Levelised cost of hydrogen (LCOH)*

The Levelised Cost of Hydrogen (LCOH) is the discounted lifetime cost of building and operating a hydrogen production asset, expressed as a cost per energy unit of hydrogen produced (£/MWh_{th} HHV) (Hydrogen production costs 2021). It covers all costs to the producer including, CAPEX, OPEX, fuel, waste disposal and financing costs. LCOH is expressed as a net present cost, as shown by Eq. (5). Although LCOH cannot be used to establish business model strike prices, it is considered a viable metric for comparing different hydrogen generation technologies. The economic values used to develop the LCOH model are detailed in Table 2.

$$LCOH = \frac{NPV\ Costs}{NPV\ Hydrogen} \tag{5}$$

$$NPV\ Costs = \sum_{i=1}^{n} \frac{CAPEX_i + OPEX_i - Power\ Generation\ Revenue_i}{(1 + Discount\ Rate)^i} \tag{6}$$

$$NPV\ Hydrogen = \sum_{i=1}^{n} \frac{Hydrogen\ Production_i}{(1 + Discount\ Rate)^i} \tag{7}$$

$(n = time\ period)$

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

Table 5
Operational cost estimates.

Item	Zero residual emissions (M£)	5% residual emission (M£)	10% residual emission (M£)
Net annual OPEX	499	481	472
Natural Gas	371	361	357
Solvent Regeneration	50	46	43
Fixed OPEX	56	54	52
CO_2 Transport and Storage (T&S)	44	41	38
Debt Financing	2	2	2
Power Generation Revenue	−25	−22	−21

2.7. *Capital expenditure (CAPEX)*

A detailed multi-level factorial cost model, shown in Eq. (13) and provided in the supplementary material is used to calculate the required CAPEX, with an estimated accuracy of +35%/−15% corresponding to an American association of cost engineers (AACE) Class 4 estimate (Christensen, 2005), which is considered sufficient for project feasibility studies. The capital cost estimates are presented in Table 4 and details of the parameters for the cost model and there sources are presented in Table 2.

Engineering, Procurement and Construction (EPC) cost for an SMR is estimated using Eq. (8) with the reference cost obtained from a report issued by the BEIS (Hydrogen production costs 2021). Hydrogen production capacity is used as the scaling parameter and the scaling exponent is set at 0.79 as per Sinnott and Towler (Sinnott and Towler, 2020).

$$SC_t = RC * \left(\frac{SP}{RP}\right)^{Exp} * \left(\frac{CEPCI_t}{CEPCI_{ref}}\right) * \left(\frac{ICCI_t}{ICCI_{\ ref}}\right) * \left(Exchange\ Rate_{ref}\right) \tag{8}$$

Where *Exp* is the Scaling Exponent, *RC* is the reference plant cost, *RP* is the reference plant parameter, *SP* is the scaled plant parameter, *SC* is the scaled plant cost, CEPCI is the chemical engineering plant cost index, ICCI is the international construction cost index, T is the current year and ref is the reference year.

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, Woods (Donald, 2007) and the available FEED studies (Elliott, 2022; Bechtel 2019; Bechtel 2018) and converted to UK 2023 prices using the chemical engineering plant index (CEPI) (THE CHEMICAL ENGINEERING 2022), historical currency exchange rates and the international construction cost index (ICCI) (Arcadis 2020). A physical module cost (PMC) for each piece of equipment is evaluated using Eq. (9) while the total plant cost (TPC) is the summation of the PMC for each piece of equipment (Eq. (10)).

$$Physical\ Module\ Cost\ (PMC) = SC * \left(1 + f_{installation} + f_{tax\ \&\ freight}\right) + I \tag{9}$$

$$Total\ Plant\ Cost\ (TPC) = \sum PMC \tag{10}$$

Where $f_{installation}$ is the installation cost relative to the equipment purchase cost and specific to each type of equipment however it is usually between 2 and 7. $f_{tax\ \&\ freight}$ is the cost of delivery of a piece of equipment to site and is set at 10% of the purchase cost and I is the specific equipment instrumentation cost.

Factors for civil works, utility installation, electrical equipment and project management are estimated from the front end engineering design (FEED) studies available in the public domain (Elliott, 2022; Bechtel 2019; Bechtel 2018) and applied to the TPC for each major PCC plant item to get a Bare Module Cost (BMC) as shown in Eq. (11).

$$Bare\ Module\ Cost\ (BMC) = \left(1 + f_{project\ management}\right) * \left[\left(1 + f_{civil} + f_{utility} + f_{electrical}\right) * TPC\right] \tag{11}$$

Table 6
Sensitivity study parameters and ranges.

Item	Lower	Base	Upper
Natural Gas price (HHV)	5 £/MWh	28£/MWh	100 £/MWh
Electricity Price	0 £/MWh	105 £/MWh	200 £/MWh
CAPEX	−15%	As evaluated	+35%
OPEX	−30%	As evaluated	+30%
Discount Rate	5%	7.8%	20%
Carbon Price	0 £/tCO$_2$	0 £/tCO$_2$	200 £/tCO$_2$
Load Factor	0.6	0.95	1
T&S Cost	10 £/tCO$_2$	19 £/tCO$_2$	50 £/tCO$_2$
DACCS Price	100 £/tCO$_2$	300 £/tCO$_2$	1000 £/tCO$_2$
Natural Gas Supply Chain Emissions	0.2%	1.04%	3.0%

A 3% contractor fee as per Woods (Donald, 2007) and a 10% contingency consistent with common FEED study practice is applied to the BMC to get the total EPC cost.

$$\text{Engineering, Procurement and Construction (EPC)} = \left(1 + f_{contractor\ fee} + f_{contingency}\right) * BMC \tag{12}$$

Total CAPEX burden is then found by adding owner's, and regulatory costs to the total EPC value while including any additional capital requirement for connections to natural gas, CO_2 and electricity networks, start-up/spares costs, consultancy fees and interest repayments accrued during the construction period as per Eq. (13). As working capital is assumed to be recovered at the end of the operational life of the plant, it is not included in the total CAPEX burden. The debt associated with working capital is serviced annually for the duration of the plant life wherein it is released.

$$CAPEX = (EPC_{SMR} + EPC_{PCC}) * \left(1 + f_{owners} + f_{regulatory} + f_{spares\ \&\ start-up} + f_{connections} + f_{consultancy}\right) + Interest\ during\ construction \tag{13}$$

A breakdown of the PCC EPC costs for both the zero, 5% and 10% residual emissions cases are shown in Table 3 while Table 4 shows the total plant CAPEX burden.

3. Operational expenditure (OPEX)

OPEX comprises any ongoing costs related to normal operations of a plant and includes fuel, consumables, labour and maintenance costs as well as the cost of CO_2 transport and storage, emissions penalties (i.e. carbon tax), waste disposal and insurance or taxes. Any revenue from the sale of power is subtracted from total annual costs to provide a net annual cost value as per Eq. (6). An MEA consumption rate of 2 kg/tCO$_2$ is assumed based on estimations by Elliott et al. (Elliott et al., 2021) for a desorber operating at 2.4 bar and 130 °C, resulting in an annual solvent replacement cost of 3.8–4.3M£ while the price of disposing of the reclaimer bottoms is assumed to be 500£/t, adding an additional 2.0–2.3M£ per annum. It is assumed that 60 permanent staff members will be required to maintain and operate the SMR and PCC plant. An additional 1.5% of the EPC cost is assumed to cover annual insurance, taxes and overheads. The natural gas prices presented in this report are based off projected wholesale fossil fuel costs for 2025, available at (Fossil fuel price assumptions 2022), while the average electricity price is taken as the levelised cost of electricity for zero residual emission electricity produced using a combined cycle gas turbine (Mullen and Lucquiaud, 2022) using the same natural gas price of 28£/MWh$_{th}$ HHV and serves as a best estimate for long term uninflated values. Table 5 presents the results for the OPEX analysis; no carbon price is assumed in this instance.

3.1. Cost of CO_2 avoided (CCA)

The cost of CO_2 avoided (CCA) expresses a technology-independent cost of reducing CO_2 emissions from a given activity per tonne of CO_2e (£/tCO$_2$e).

The CCA for the end use cost of fuel switching from natural gas to hydrogen i.e. gas-to-blue is described by Eq. (14) (Rehl and Müller, 2013).

$$Cost\ of\ CO_2Avoided_{gas\ to\ blue} = \frac{LCOH - Natural\ Gas\ Price}{Natural\ Gas\ lifecycle\ emissions - \ Hydrogen\ lifecycle\ emissions} \tag{14}$$

Where LCOH is the levelised cost of hydrogen (£/MWh$_{th}$), natural gas price is the wholesale cost of natural gas (£/MWh$_{th}$) while hydrogen and natural gas emissions is the lifecycle CO_2 equivalent emissions resulting from the supply of one unit of heat via hydrogen or natural gas respectively (tCO$_2$e/MWh$_{th}$). For the purpose of this work gas-to-blue CCA is reported on both a HHV and LHV as the appropriate metric is dependant on end use of the H_2 product. A large portion of the applications for fuel switching from natural gas to hydrogen require high grade heat, i.e. open cycle gas turbines, furnaces or domestic/industrial boilers, where the latent heat of H_2O vaporisation is typically not recovered. For applications such as this a CCA reported on a LHV basis

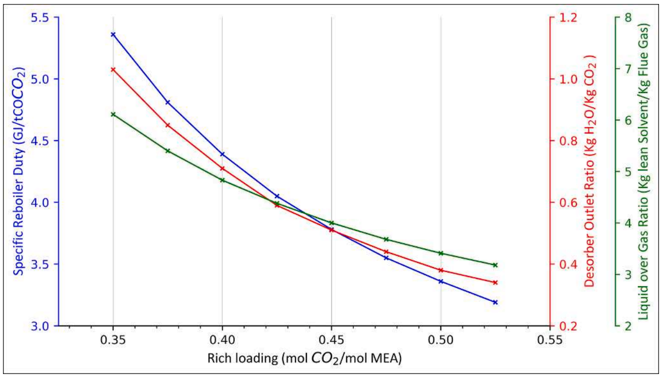


Fig. 4. Effect of increasing rich loading on specific reboiler duty for a constant lean loading of 0.16 mol CO_2/mol MEA and 20% v/v flue gas CO_2 concentration and 100% fossil CO_2 capture.

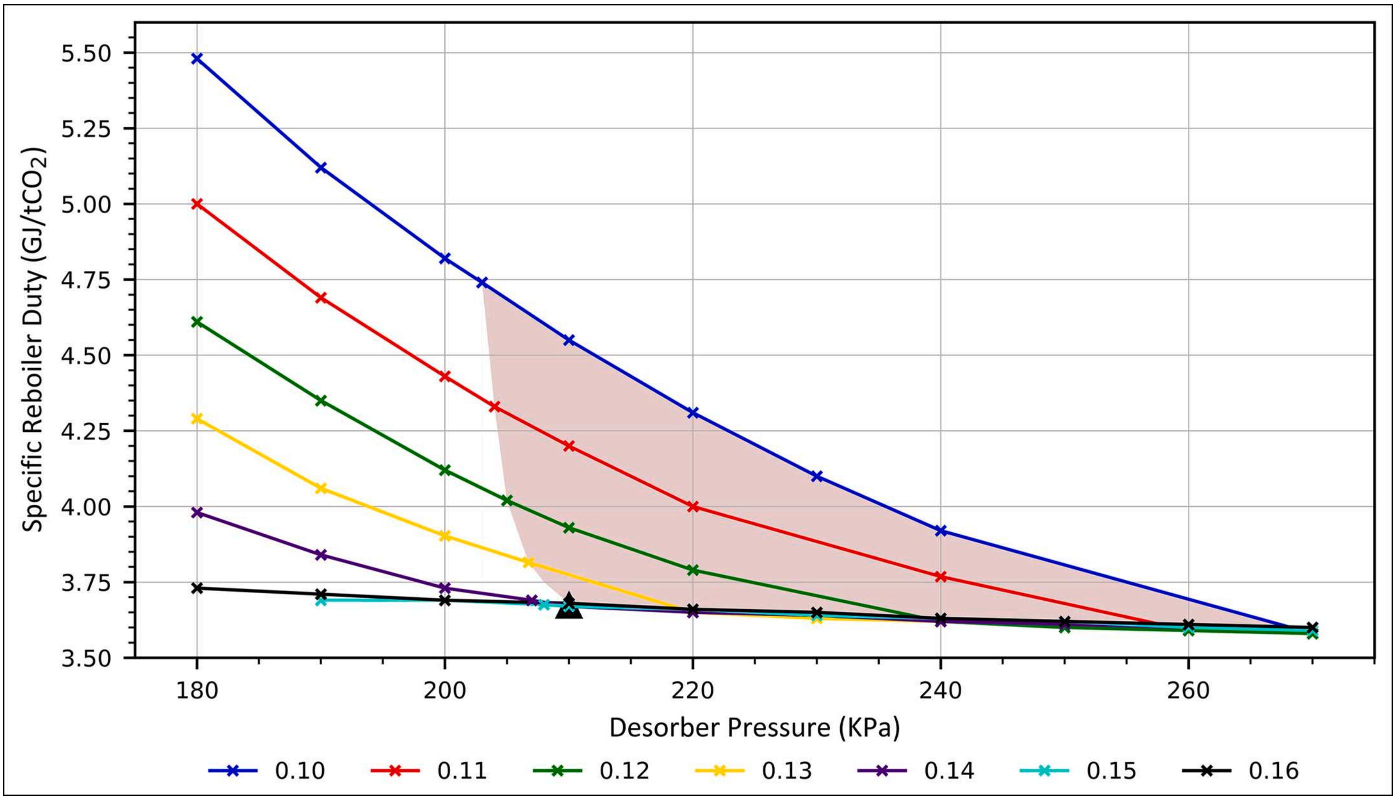


Fig. 5. Effect of desorber operating pressure on the specific reboiler duty for a range of lean solvent loadings (mol CO_2/mol MEA) for a rich loading of 0.47 mol CO_2/mol MEA. Shaded zone represents operational range where reboiler temperature is above 125 °C.

may be more representative, however for applications where the latent heat of H_2O vaporisation is recovered such as condensing boilers HHV may be more appropriate.

3.2. Levelised cost of capture (LCOC)

The Levelised Cost of Capture (LCOC) is the discounted lifetime cost of building and operating a CO_2 capture asset and is expressed as a cost per tonne of CO_2 captured £/tCO_2 and defined by Eq. (15) (Review of next generation 2022). The LCOC is typically presented without including transport and storage costs, and as such that convention is maintained in this study.

$$LCOC = \frac{NPV\ Costs}{Total\ discounted\ lifetime\ CO_2 captured} \tag{15}$$

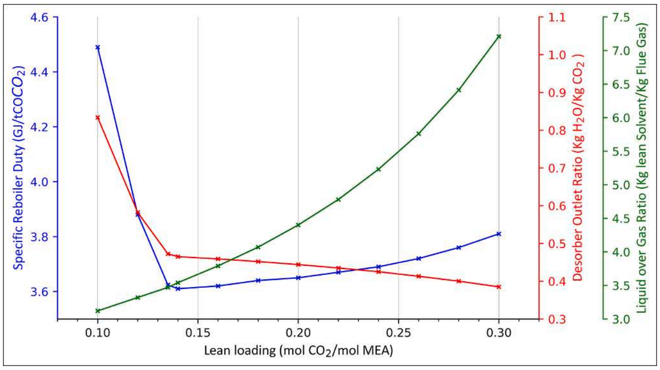


Fig. 6. Reboiler Duty, L/G and desorber outlet ratio for constant rich loading of 0.47 mol CO_2/mol MEA and desorber operating pressure of 210kpa.

3.3. Sensitivity study

Upper and lower estimates of ten key operating and economic parameters are investigated and the impact on the LCOH presented. Table 6 details the bounds of the sensitivity study.

4. Results and discussion

4.1. Zero residual emissions

The results presented in this section show that, for the assumptions used, the ultra-high capture fractions required to achieve zero residual emissions from SMR+CCS plants can be achieved economically through optimisation of the post combustion CO_2 capture plant design. The thermal efficiency of H_2 production decreases by just 1.9 percentage points while total plant CAPEX and OPEX are increased by 5.3% and 4.3% respectively, relative to the 5% residual emissions case.

4.2. Design of a post combustion CO_2 capture plant for ultra-high capture fraction

Optimisation of the post combustion CO_2 capture plant design for ultra-high capture fractions necessitates using lean solvent loadings, in this case 0.16 mol CO_2/mol MEA, lower than those typically reported in the literature for a 90% capture fraction (circa 0.2–0.25 mol CO_2/mol MEA). This ensures that the driving force for CO_2 absorption at the top of the absorber remains sufficient to allow effective utilization of the available absorber packing when intercooling is also employed. Under these conditions an additional 6 m of absorber packing is also required to reach reasonably high rich loadings, limiting the increase in thermal energy requirement to just 1.6%. A strong relationship between desorber pressure and specific reboiler duty is observed, indicating that increased desorber pressure, and hence also increased reboiler

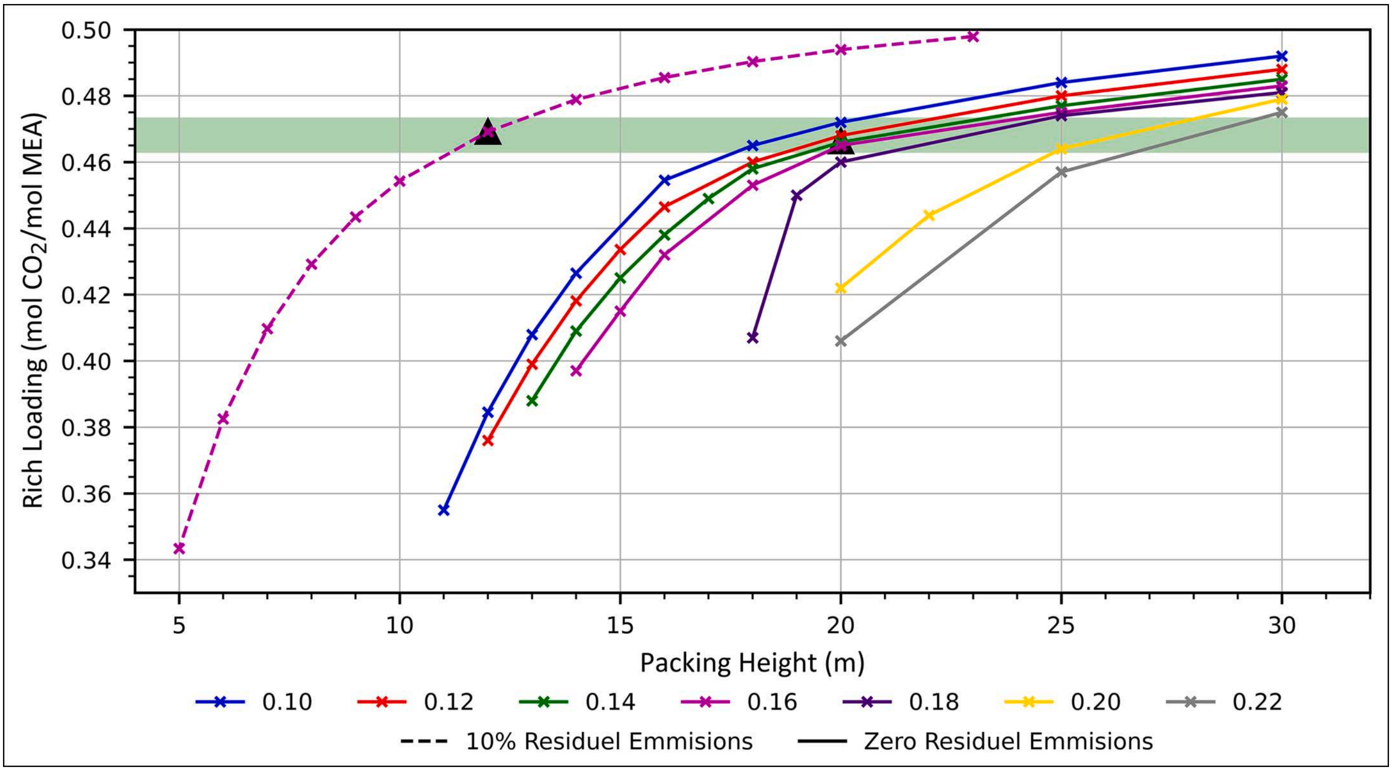


Fig. 7. Effect of packing height on rich solvent loading for a range of lean loadings. The green horizontal bar represents 86.5–88.5% of rich loading equilibrium while the black triangles represent chosen design points. Flue gas CO_2 concentration is 20 vol%.

Table 7
Design and operating parameters of the CO_2 capture plant.

Parameter	Unit	Zero residual emissions	5% residual emission	10% residual emission
§Flow Rate (Flue Gas)	*kg/s*	275	262	256
Inlet Temperature	°*C*	40	40	40
CO_2 Concentration	*Mole Frac*	19.4	19.9	20.2
Absorbers	–	1	1	1
Packing Stages	–	2	2	2
Packing Height	*m*	20	14	12
Diameter	*m*	12	12	12
Packing Volume	m^3	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
Lean Loading	*mol* CO_2/ *mol MEA*	0.16	0.16	0.16
Rich Loading	*mol* CO_2/ *mol MEA*	0.466	0.469	0.469
Operating Pressure	*KPa*	210	210	210
Reboiler Temperature	°*C*	125	125	125
Specific Reboiler Duty	*GJ/t*CO_2	3.67	3.62	3.60

temperatures, may be required at low lean loading levels in order to limit the increase in specific reboiler duty.

Fig. 4 shows how, for a constant lean loading and CO_2 capture fraction, an increasing rich loading at the bottom of the absorber decreases specific reboiler duty, this analogous of increasing the total packing height. As rich loading increases the CO_2 absorbed per unit of

Table 8
Performance assessment of a SMR plant with zero, 5% and 10% residual emission PCC.

Parameter	Unit	Zero residual emissions	5% residual emission	10% residual emission
H_2 Export (HHV)	MW_{th}	1000	1000	1000
H_2 Export	kg/s	7	7	7
House Load	MW_e	−26	−24	−23
Net Power Output	MW_e	33	30	28
Supplementary fuel	kg/s	8.7	7.9	7.5
Feedstock	kg/s	20	20	20
Total Fuel	kg/s	29	28	28
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
Specific carbon intensity of H_2	gCO_2e/ MJ LHV	5.0	9.0	13.2

Table 9
LCOH, LCOC, CCA and CAC for Zero, 5% and 10% residual emission operation.

Case	LCOH (£/MWh_{th})	LCOC (£/tCO_2)	CCA (gas-to-blue) (£/tCO_2e) HHV	CCA (gas-to-blue) (£/tCO_2e) LHV
No CO_2 abatement	44.2	–	–	–
10% residual emissions	65.3	64.2	254.6	289.7
5% residual emissions	66.4	63.3	240.9	271.9
Zero residual emissions	68.8	64.7	237.5	265.9

Table 10
LCOH for Zero, 5%, 10% residual and no CO_2 abatement operation with a range of carbon tax applied.

Carbon Tax (£/tCO_2)	Zero residual emissions	5% residual emissions	10% residual emissions	No CO_2 abatement
0	68.8	66.4	65.3	44.2
100	68.8	67.7	67.9	69.5
200	68.8	69.0	70.6	94.7
300	68.8	70.3	73.2	119.9
500	68.8	71.6	75.8	145.2

Table 11
LCOH breakdown: No Carbon pricing.

		Zero residual emissions	5% residual emission	10% residual emission
Fuel	£/MWh_{th}	41.9	40.7	40.2
CAPEX	£/MWh_{th}	17.9	17.0	16.7
OPEX	£/MWh_{th}	6.7	6.4	6.3
CO_2 Transport and storage	£/MWh_{th}	5.3	4.9	4.6
Power Generation Revenue	£/MWh_{th}	−3.0	−2.7	−2.5
Total	£/MWh_{th}	68.8	66.4	65.3

solvent increases proportionately; this reduces the sensible heat addition required to heat the solvent to the desorption temperature decreases (see Eq. (2)) as the quantity of water contained in the solvent mixture per unit of CO_2 captured decreases. Concurrently the water VLE at the top of the desorber changes with rich loading, as represented by the H_2O to CO_2 desorber outlet ratio. This tends to further decrease the specific reboiler duty at higher rich loadings.

For a 35% wt MEA solvent and rich loading of 0.47 mol CO_2 a strong effect of varying desorber pressure, and by extension reboiler temperature, is observed for lean solvent loadings at the lower end of the investigated range 0.10 – 0.14 mol CO_2/mole MEA, as illustrated in Fig. 5. This is because the VLE of H_2O at the bottom of the desorber was found to be highly dependant on both desorber pressure and CO_2 loading, a relationship that tends to result in increased vaporisation of H_2O at lower pressures and CO_2 loadings, leading to a rise in required thermal input. This is illustrated in detail in Fig. 6 for a desorber operation pressure of 210KPa. The effect becomes less significant for lean loadings equal or higher than 0.15 mol CO_2/mol.

The 125°C solvent temperature ceiling imposed in this study limits the minimum lean loading to 0.15 mol CO_2/mole MEA at a desorber pressure of 210 KPa (as per Fig. 5) if excessive specific reboiler duties values are to be avoided; a slightly higher value of 0.16 mol CO_2/mol MEA was chosen as the default lean loading in all cases to allow for operational flexibility.

Since lean loadings in the 0.10 – 0.14 mol CO_2/mole MEA range are likely to be required to achieve economical capture of 100% of the fossil CO_2 from flue gases with lower CO_2 concentrations than SMR's, as shown by Michailos and Gibbins (Michailos and Gibbins, 2022), increased desorber pressures, and hence increased reboiler operating temperatures, will be required to limit the increase in specific reboiler duty in these applications. Such low lean loadings may also have advantages for SMR PCC, e.g. avoiding the need for intercooling or facilitating reduced packing heights. This may result in an increase in MEA consumption per tonne of CO_2 captured due to increased thermal degradation but the impact on the cost of CO_2 capture could be minimal given the MEA replacement costs of 2.3\$/tCO_2 estimated by Elliott et al. in 2021 for a desorber operating at 240KPa and 130 °C (Elliott et al., 2021)

Table 12
LCOH for various natural gas supply chain emissions rates over the range of DACCS cost estimates.

Case	Natural Gas supply chain emission rate (% fuel supplied)	LCOH £/MWh_{th} HHV (DACCS Cost = 100–1000 £/tCO_2)	Increase in fuel cost (%) (Natural gas to H_2)	CCA (HHV) (£/tCO_2e)	CCA (LHV) (£/tCO_2e)
Low	0.20%	70–79	150–182	223–272	248–300
UK	1.04%	71–86	153–208	222–302	247–332
High	3.00%	73–103	160–266	220–367	244–401

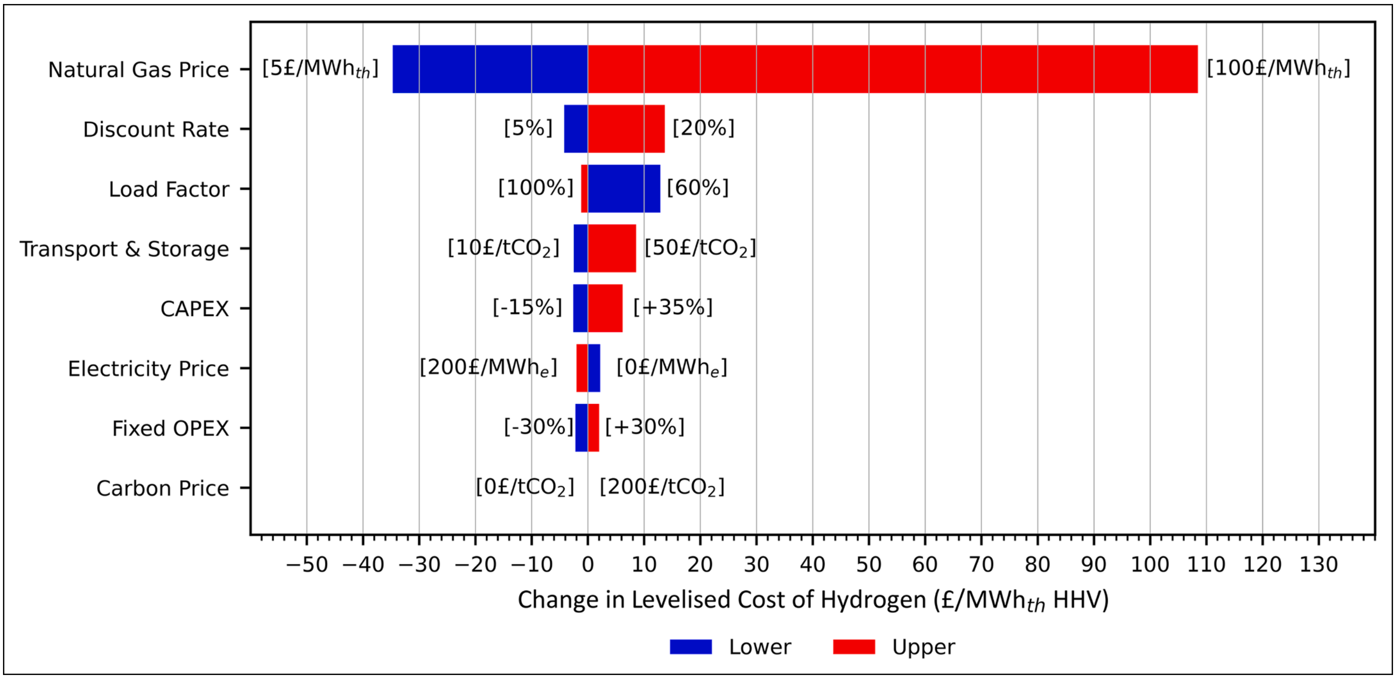


Fig. 8. Effect of techno-economic parameters on the LCOH for zero residual emission operation sensitivity study.

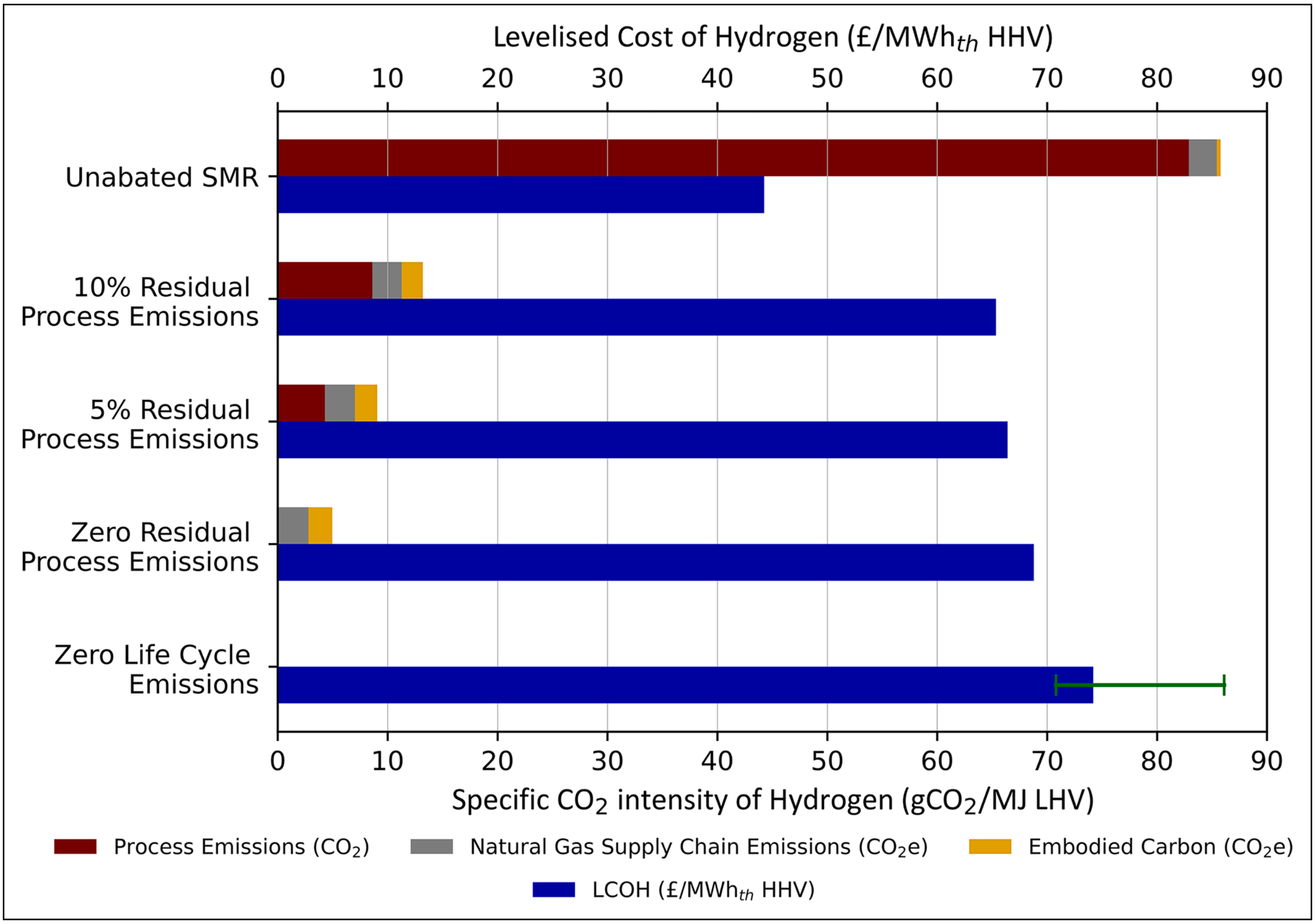


Fig. 9. CO_2 intensity of H_2 production vs LCOH for a UK case (1.04% natural gas supply chain emissions and DACCS at 100 - 1000£/tCO_2). Green bar represents the change in LCOH over the range of DACCS costs analysed.

Fig. 7 illustrates the effect of increasing the absorber packing height on rich loading over a range of solvent lean loadings for both a zero residual and 10% residual emission configurations. In general, operating at lower lean loadings and appropriately lower L/G ratios leads to reduced packing height while still achieving sufficiently high rich loadings. In this study, the packed bed CO_2 absorber is designed so that the rich solvent loading approaches 87.5% of the absolute value of solvent loading at equilibrium at the bottom of the absorber (ca. 0.47 mol CO_2/mol MEA). It is believed that this may represent a reasonable trade-off between CAPEX-related absorber packing height and OPEX-related specific reboiler duty.

The 10% residual emission case requires less packing height to achieve rich loadings of or above 87.5% of equilibrium and can do so at higher lean loadings, should that be required. This is primarily due to the increased partial pressure of CO_2 at the top of the absorber relative to the zero residual emissions case. 12 and 14m of packing is used for the 5 and 10% residual emission cases respectively and is broadly in line with what is reported in the literature. 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 (Bechtel 2018). 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 (Nexant 2016). 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.

It is worth noting that the rich loading achieved for the zero residual emissions configuration is slightly below that of the 5 and 10% residual emissions cases at 0.466 vs 0.469 mol CO_2/mol MEA. This is a marginal compromise to meet the 20 m packing height constraint discussed in section 2.1; increasing packing height further would require a third bed and impart a step change in absorber CAPEX.

Exact optimum points for absorber design are hard to quantify as they will be dependant on project specific constraints i.e. CAPEX availability, OPEX impacts, access to heat for solvent regeneration, land availability, maximum absorber height restraints etc. As such absorber design should be assed and optimised on a project specific basis.

4.3. Performance assessment of the effect of increased CO_2 capture fractions on an SMR plant with PCC

Table 7 provides a summary of the design and operating parameters of the PCC plant for the 5%, 10% and zero residual emissions configurations.

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 minimal 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

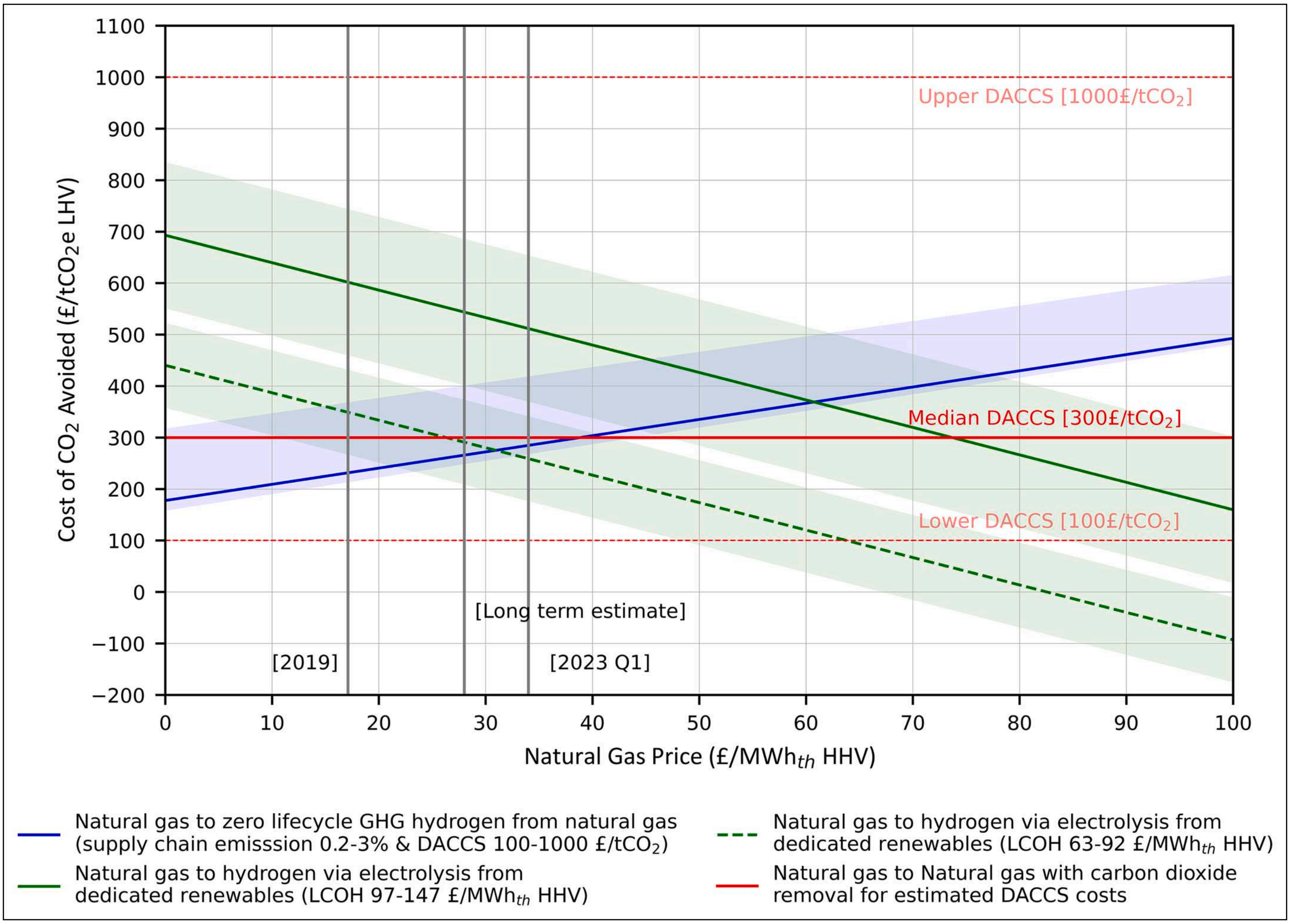


Fig. 10. A cost of CO_2 avoided comparison of fuel switching options for a range of natural gas prices on a LHV basis, including 2019, 2023 and long term estimate prices. Shaded regions indicated upper and lower bounds.

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 increases proportionately in order to supply sufficient steam for solvent regeneration. This leads to the increased flue gas flow rate and 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 7.

Table 8 shows the performance of the SMR operating with zero residual emissions compared to 5% and 10%.

The additional steam for solvent regeneration required to increase the CO_2 capture fraction to zero residual emissions results in an increase in the consumption of natural gas in the burner, causing 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.

4.4. Levelised cost of hydrogen production

An economic analysis predicting the levelised cost of hydrogen (LCOH), levelised cost of capture (LCOC) and cost of CO_2 avoided (CCA) is shown for zero, 5% and 10% residual emissions operation of a 1000 MW capacity SMR when no carbon pricing is applied, the results are detailed in Table 9 and Table 11. Under these conditions zero residual emission operation is shown to be just 5.3% and 3.6% more costly than hydrogen produced with 10% and 5% residual emissions respectively. When a carbon tax is introduced, as in Table 10, the situation changes, as cases with residual emissions will be penalised when a carbon tax is present a breakeven point occurs. Table 10 shows that for a carbon tax of just 100£/tCO_2 all CCS enabled hydrogen production is more cost effective than an unabated SMR. At 200£/tCO_2 zero residual emission operation is the most cost effective option of the configurations analysed.

However, the primary reason for the mass production of hydrogen is to decarbonise a carbon intensive energy vector, likely natural gas, and as such cost is not the only metric of value. The cost of CO_2 (CCA) avoided allows for the direct comparison of H_2 production methods per unit of CO_2 avoided, it can be used to form a holistic view of all options available to determine the least cost option for moving away from a carbon intensive energy vector. When the CCA (direct emissions only) of switching from natural gas to hydrogen is considered (assuming combustion as the final use), zero residual emission production was found to be the lowest cost option with a value of 238 £/tCO_2e on a HHV bases and 266 £/tCO_2e on a LHV bases and a natural gas price of 28£/MWh_{th} (HHV), this conclusion remains true for all natural gas prices analysed as part of this study.

Table 9 shows that the LCOC and CCA follows a non-linear relationship to capture fraction; this is tentatively attributed to non-linear CAPEX relationships, increase fuel requirements and varying specific

Table 13
Detailed Process parameters for a Zero Direct Emission SMR.

Parameter	Unit	Value
Reformer Operating Pressure	Bar	32
Reformer Reaction Temperature	°C	913
Methane Conversion Efficiency (Reformer)	%	85
Steam to Carbon Ratio (Reformer)	–	2.5
Pre-Reformer Reaction Temperature	°C	500
Carbon Monoxide Conversion Efficiency (WSR)	%	72
WSR Reaction Temperature	°C	410
Flue gas flow rate	Kg/s	275
Flue gas CO_2 concentration	% Vol	20
Turbine Inlet Pressure	Bar	42
Turbine Outlet Pressure	Bar	3.7
Turbine Inlet Temperature	°C	400
Turbine Isentropic Efficiency	%	90
Pump Isentropic Efficiency	%	70
Fan Isentropic Efficiency	%	70
Generator Efficiency	%	99
Mechanical efficiency (Turbine)	%	99
Mechanical efficiency (Pump & Fan)	%	95
CW water supply temperature	°C	8
CW return temperature	°C	25
Economizer steam side ΔP	%	1
Superheat steam side ΔP	%	2
Heat Recovery gas side ΔP	%	0.1
Pre-reformer ΔP	%	7
Reformer ΔP	%	10
WSP ΔP	%	5
Minimum Pinch Temp (Heat recovery)	°C	10
Minimum Flue gas Oxygen concentration	% vol	1
Solvent (MEA composition)	% wt	35
Specific reboiler duty	GJ/tCO_2	3.67
Lean/Rich HX minimum approach temperature	°C	10
Minimum Solvent return temperature	°C	35
Minimum Intercooler return temperature	°C	25
Absorber Flooding point	%	80
Lean Solvent loading	Mol/Mol	0.16
Desorber operating pressure	KPa	210
Structured Packing ΔP	mbar/m	2
Desorber packing	–	MELLAPAK 252Y
Absorber packing	–	MELLAPAK 252Y
Maximum Reboiler Temperature	°C	125
Rich Loading Target	% VLE	85
Gas Mixture Thermodynamic Model	-	Peng-Robinson
Steam/Water Thermodynamic Model	-	IAPWS-95

Table 14
Natural Gas composition.

Species	% vol	Species	% vol
Methane	89.00	Propane	1.00
Ethane	7.00	Nitrogen	0.89
Carbon Dioxide	2.00	Butane	0.11

reboiler duty as detailed in Table 11.

5. Sensitivity study

The results from the LCOH sensitivity study completed using the range of parameters described in Table 6 are shown in Fig. 8; each parameter is varied in turn while the remaining parameters are kept constant. The LCOH is shown to be highly sensitive to the natural gas price and moderately sensitive to discount rate and load factor. Variation in CAPEX has a less prominent effect with a −15% to +35% swing resulting 3.1% variance in LCOH.

5.1. *Compensating for supply chain emissions & embodied carbon*

As shown in Table 9, In all cases the CCA of zero residual operation is found to be the lowest cost decarbonisation option for SMRs, as a result the authors consider 100% fossil CO_2 capture in the CO_2 capture process as likely to be the preferred pathway to achieving zero life cycle CO_2 emission hydrogen from natural gas in SMRs. The results presented in this section consider a zero residual emission SMR as the hydrogen production method.

Given the uncertainty in both future DACCS prices and natural gas supply chain emissions, LCOH values for three cases are presented in Table 12 for zero life cycle CO_2 emissions hydrogen production using steam methane reforming with zero residual emission CO_2 capture.

The first case is an optimistic outlook wherein global natural gas supply chain emissions fail in line with current best practices and achieve an average global rate of 0.2%. The second cases represents a mid-range estimate where current UK natural gas supply chain emission rates are used (1.04%). The final case represents a pessimistic outlook where global natural gas supply chain emissions, at 3%, are twice the median value of 1.5% assumed by Bauer et al. Total embodied carbon is estimated at 1.4MtCO_2e, which is offset via additional DACCS. Construction emissions are offset during the construction period and CCS fugitive emissions are offset annually. Estimations for the LCOH of zero lifecycle CO_2 emissions hydrogen production range from 70£/MWh_{th} HHV on the low end to 103£/MWh_{th} HHV in the most pessimistic scenario, representing a 2–50% increase in LCOH as a result of offsetting supply chain and embodied carbon. For the UK case with a mid-range estimate for the price of DACCS of 300/tCO_2 the LCOH of zero lifecycle CO_2 emissions hydrogen is 74£/MWh_{th} HHV, an 8% increase.

Fig. 9 shows the life cycle CO_2e emissions and associated LCOH for five representative cases for large scale hydrogen production in the UK using SMR technology. As would be expected the cost of H_2 production increases as the degree of decarbonisation increases, this is primarily due to the increase CAPEX and OPEX of the CCS plant on an absolute basis. However when considered on a specific decarbonisation basis, with metrics such as CCA, it is clear that, for the assumptions detailed in this paper, the least cost decarbonisation pathway trends towards increasing CO_2 capture rates.

5.2. *Sensitivity study*

Both natural gas emission rates and DACCS prices are expected to vary both spatially and temporally, highlighting the importance of case by case assessments, spatially due to varying natural resources, financial incentives and infrastructure between regions, and temporally as pledges to reduce natural gas supply chain emissions come into effect and DACCS technology matures.

For fuel switching to hydrogen to be economical the LCOH must be such that the CCA (as per Eq. (14)) is lower than the current cost of DACCS, otherwise, recapturing natural gas emissions via DACCS is the least cost option. In practice, as the presented values include only the relative fuel supply costs, the breakeven LCOH that would make hydrogen competitive with direct natural gas use with DACCS is somewhat lower than shown in Fig. 10. This is due to the costs of conversion of the distribution system and end use facilities from natural gas to hydrogen. However a detailed examination of this is beyond the scope of this paper.

Fig. 10 illustrates how the CCA of fuel switching from natural gas to blue hydrogen is sensitive to natural gas prices. Should natural gas prices return to those seen prior to 2020 at circa 17£/MWh_{th} HHV (50p/therm) the CCA for switching to zero lifecycle emission hydrogen from natural gas would reduce to below 213–368£/tCO_2 on a LHV basis for the UK. However, should that not be the case and natural gas prices remain at the levels encountered at the time of writing (i.e. the first half of 2023) of 34.1 £/MWh_{th} HHV (100p/therm (Ofgem 2022)) the CCA for gas-to-blue switching would be 268–419£/tCO_2. A long term estimate for natural gas wholesale price of 28£/MWh_{th} HHV (82p/therm) (Review of next generation 2022) results in a CCA of 248–401£/tCO_2.

Of note is the fact that no allowance is made for cost reductions in the post combustion carbon capture system over time in this analysis, and

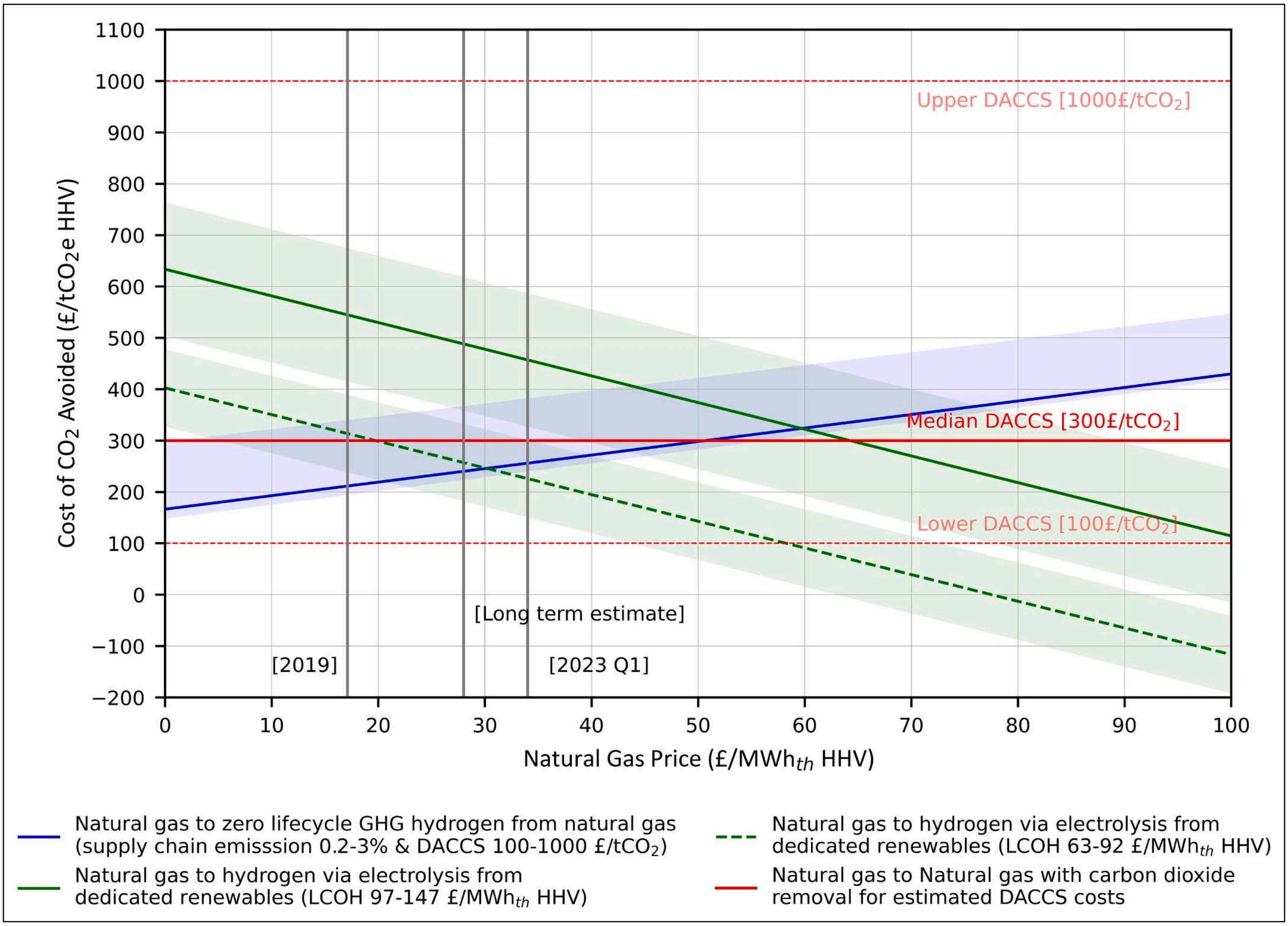
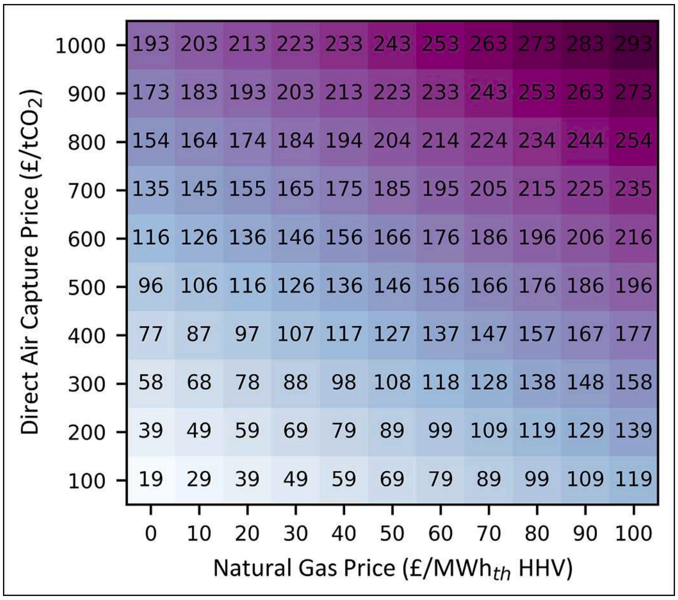


Fig. 11. LCOH of Zero life cycle emission H_2 vs Natural Gas Emissions rates and DACCS Cost.



	0	10	20	30	40	50	60	70	80	90	100
1000	193	203	213	223	233	243	253	263	273	283	293
900	173	183	193	203	213	223	233	243	253	263	273
800	154	164	174	184	194	204	214	224	234	244	254
700	135	145	155	165	175	185	195	205	215	225	235
600	116	126	136	146	156	166	176	186	196	206	216
500	96	106	116	126	136	146	156	166	176	186	196
400	77	87	97	107	117	127	137	147	157	167	177
300	58	68	78	88	98	108	118	128	138	148	158
200	39	49	59	69	79	89	99	109	119	129	139
100	19	29	39	49	59	69	79	89	99	109	119

Fig. 12. A cost of CO_2 avoided comparison of fuel switching options for a range of natural gas prices on a LHV basis, including 2019, 2023 and long term estimate prices. Shaded regions indicated upper and lower bounds.

that the costs presented here represent first of a kind status. Learning as deployment increases and the technology approaches N_{th} of a kind status will reduce the CCA of fuel switching. Estimated LCOH values in 2025 (97–147£/MWh_{th} HHV) and 2050 (63–92£/MWh_{th} HHV) for hydrogen production via electrolysis with dedicated renewable energy (green hydrogen) published by BEIS (Hydrogen production costs 2021) are shown for illustrative purposes as the shaded green regions in Fig. 10. Implicit in the assumption of the above LCOH values for green hydrogen is that no capacity to export power to the national grid is present. This means the value of that power is independent from the national grid and as such is unaffected by peaks in the wholesale electricity price. Should that not be the case the LCOH of green hydrogen would be linked to the wholesale electricity price, and by extension, given the current market structure, the wholesale natural gas price. All other factors being equal this would result in an increase in the CCA of green hydrogen. Fig. 10 also assumes zero life cycle emissions from green hydrogen production. This will likely result in an under estimation of the CCA, quantification of the life cycle emissions from green hydrogen production is, however, beyond the scope of this paper.

6. Conclusions

We show, for the first time, that transitioning to zero life-cycle CO_2 emission hydrogen production from natural gas involves a minor increase in costs relative to the traditionally assumed CO_2 capture fraction of 90–95% and a reduction in the cost of CO_2 avoided (CCA). This leads the authors to the novel conclusion that 100% fossil CO_2 capture in the

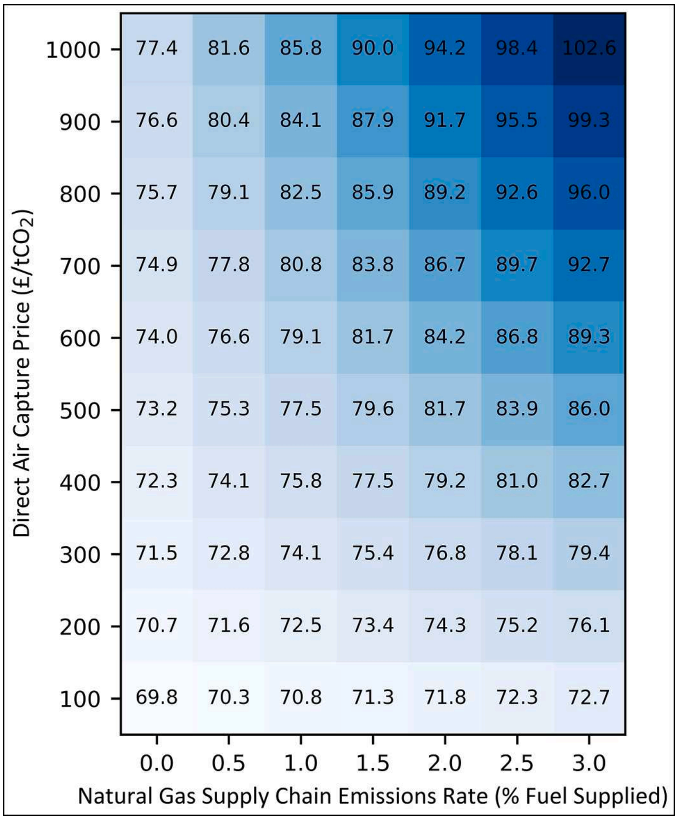


Fig. 13. Maximum economic levelised cost of net zero hydrogen for a range of natural gas and DACCS prices. Analyses is independent of hydrogen production method.

CO_2 capture process is likely to be the preferred pathway to achieving zero life cycle CO_2 emission hydrogen from natural gas in SMRs

The first step is to achieve zero residual emissions from the production process. This would require the industry and blue hydrogen project developers to design plants that are capable of capturing 100% of fossil CO_2 emissions, a technological requirement believed to be, based on the studies referenced and results presented in this paper, achievable with commercially available technology and open art solvents. We combine insights from our process modelling with a multi-level financial assessment tool developed to inform a techno-economic model describing a steam methane reformer (SMR) equipped with post-combustion CO_2 capture capable of 100% of fossil CO_2 capture.

The PCC plant design proposed in this study differs from conventional PCC design only due to the increase in packing height (14 m to 20 m) and reduced solvent lean loadings (0.16 from circa 0.20–0.25 mol CO_2/mol MEA). This is necessary to limit CO_2 capture thermal energy use to a minimal increase of 1.6% per tonne of CO_2 captured. The additional gas input needed to increase CO_2 captured per unit of H_2 produced lowers hydrogen production efficiency by 1.9% point HHV, from 68.7 to 66.8%. This, coupled with the use of an open art solvent (35%wt MEA), leads to the conclusion that zero residual emission operation of an SMR can be achieved using currently available technology and at a much lower efficiency impact than previously thought.

Additionally, it is estimated that capital and operating costs increase by, respectively, 5.3% and 4.3% for a 1000 MW HHV hydrogen facility, compared to hydrogen produced with 5% residual CO_2 emissions. The Levelised Cost of Hydrogen with zero residual emissions is 69£/MWh_{th} HHV (2.7£/kg) for 2023 UK capital costs and a long term natural gas price of 28 £/MWh_{th} HHV, this represents a 4% increase in the LCOH relative to operating with 5% residual emissions and a 55% increase from an SMR with no CO_2 abatement.

The second step towards zero-carbon hydrogen requires using carbon-dioxide removal (CDR) technologies to recapture greenhouse gas emissions across the whole hydrogen supply chain, including emissions associated with the supply of natural gas and embodied CO_2 emissions in the construction and operation of the SMR and the CCS infrastructure. Recent pledges at COP26 to reduce global methane emissions by 30% by 2030 are a partial step to make blue hydrogen more climate compatible, but current best practices will need to be implemented globally to reduce global average emission rates in the natural gas supply chain from 1.5% to 0.2% or below.

At 0.2% supply chain methane emission rate, and with a cost of DACCS with geological CO_2 storage of between 100 and 1000 £/tCO_2 (based on the lower and upper end of long-term estimates in the literature), the Levelised Cost of Hydrogen for an assumed natural gas cost of 28 £/MW_{th} HHV and electricity price of 105£/$MWh_e$with zero life cycle greenhouse gas emissions ranges from 70 to 79£/MWh_{th} HHV (2.8 - 3.1 £/kg). This is well within the range of the cost estimates for other forms of low carbon hydrogen production in the UK, with production costs for electrolysis with dedicated renewable generation estimated at 97–147 £/MWh_{th} HHV (3.8 – 5.8£/kg) for projects commissioning in 2025, falling to 63–92£/MWh_{th} HHV (2.5 - 3.6£/kg) in 2050 (Hydrogen production costs 2021).

DACCS costs have been identified as a factor in net-zero hydrogen production but they also determine whether or not it is desirable to use hydrogen in preference to natural gas. As Fig. 10 shows, at the median DACCS cost of 300 £/tCO_2 the greater natural gas use involved in blue hydrogen production means that gas+DACCS is competitive at natural gas prices above circa 40 £/MWh_{th} HHV. However, because system and end use conversion costs have not been included, the actual gas price that would make blue hydrogen unattractive at this DACCS cost would be somewhat lower. At the minimum DACCS cost considered, 100 £/tCO_2, blue hydrogen is never competitive at any gas price. However no allowance has been made for capital and operational cost reductions in the blue hydrogen production process due to learning, with assumed learning being a factor in the lower DACCS cost estimates.

We note that analogous studies of methods to achieve zero-carbon hydrogen using other hydrogen production technologies could also be undertaken and that these might offer advantages over the SMR+CCS route. It is, however, a significant finding that at least one zero-carbon hydrogen route has been demonstrated and that this is for the SMR+CCS route, which, anecdotally, is often currently assumed to offer inferior capture performance to autothermal reactor (ATR) routes. It is our hope that this study can form a basis for future research and policy decisions and help inform a more rounded discussion on hydrogen and the energy system as a whole.

Finally, the authors would like to note that this study serves not as an advocation for blue hydrogen over green hydrogen, or even for the propagation of a hydrogen economy. Rather it is an attempt to provide much needed balance to the discussion. If indeed society decides to progress the hydrogen economy, what truly matters is that this is achieved at the lowest societal and environmental cost possible. To this end we believe that blue hydrogen can play an integral part in achieving this goal; with crucial emphasis on *part*. What is becoming abundantly clear is that an equitable and timely transition to a net zero GHG economy will require every viable lever available to us, regardless of shade.

CRediT authorship contribution statement

Daniel Mullen: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization, Data curation. **Laura Herraiz:** Writing – review & editing, Investigation. **Jon Gibbins:** Writing – review & editing, Investigation. **Mathieu Lucquiaud:** Conceptualization, Supervision, Project administration, Writing – review & editing, Investigation, Resources, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

I have attached the CAPEX model as supplementary information, any additional data required can be made avalible.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.ijggc.2023.103904.

Appendix A

Table 13.
Table 14.

Appendix B

Fig. 11.
Fig. 12 and Fig. 13.

References

Alcalde, J., Flude, S., Wilkinson, M., Johnson, G., Edlmann, K., E Bond, C., et al., 2018. Estimating geological CO2 storage security to deliver on climate mitigation. Nat. Commun. 9 https://doi.org/10.1038/s41467-018-04423-1.

Arcadis, 2020. International Construction Costs 2020. The Organisation. https://connect.arcadis.com.

Aspentech. ASPEN PLUS V10 [internet]: the orginisation; 2022 [Available from: https://www.aspentech.com/en/products/engineering/aspen-plus.

Assessing the Cost, 2018. Reduction Potential and Competitiveness of Novel (Next Generation) UK Carbon Capture Technology. Amec Foster Wheeler Group. https://www.gov.uk. Report No.: 13333-8820-RP-001.

Bauer, C., Treyer, K., Antonini, C., Bergerson, J., Gazzani, M., Gencer, E., et al., 2022. On the climate impacts of blue hydrogen produciton. Sustain. Energy Fuels 6. https://doi.org/10.1039/D1SE01508G.

Bechtel, 2018. Retrofitting an Australian brown coal power station with post-combustion capture - a conceptual study. Bechtel Infrastruct. Power Corp.

Bechtel. CO2 capture facility at kårstø, norway: front-end engineering and design (FEED) study report. Feed Study. GASSNOVA; 2019 3 Apprial 2019. Report No.: 25474-000-30R-G04G-00001.

Beis, 2022. British Energy Security Strategy. Prime Minister's Office, 10 Downing Street. www.gov.uk.

Braakhuis, L., Høisæter, K.K., Knuutila, H.K., 2022. Modeling the formation of degradation compounds during thermal degradation of MEA. Ind. Eng. Chem. Res. 61 (7), 2867–2881. https://doi.org/10.1021/acs.iecr.1c04496.

Cain, M., Lynch, J., Allen, R., Fuglestvedt, M.S., Frame, J.J., Macey, D.H., 2019. A. Improved calculation of warming-equivalent emissions for short-lived climate pollutants. Clim. Atmos. Sci. 2 https://doi.org/10.1038/s41612-019-0086-4.

Ccsi. WELCOME TO THE CARBON CAPTURE SIMULATION FOR INDUSTRY IMPACT (CCSI2) [Internet]: the Orginisation 2022 [Available from: https://www.acceleratecarboncapture.org/.

Christensen, P.D., Larry. cost estimate classification system – as applied in engineering, procurement, and construction for the process industries aace; 2005. Report No.: 18R-97.

Collodi, G., Azzaro, G., Ferrari, N., 2017. Techno-Economic Evaluation of SMR Based Standalone (Merchant) Hydrogen Plant with CCS. Int. Energy Agency Greenhouse Gas.

Comparison of commercial, 2022. State-of-the-art, Sossil-Based Hydrogen Production Technologies. National Energy Technology Laboratory. Report No.: DOE/NETL-2022/3241.

Cop26, P. World leaders kick start accelerated climate action at COP26 ukcop26.org: COP26 Press; 2021 [Available from: https://ukcop26.org/world-leaders-kick-start-accelerated-climate-action-at-cop26/.

Curtis, C. Review of Best Available Techniques (BAT) for Hydrogen Production from Methane and Refiner Fuel Gas with Carbon Capture. Costain; 2022. Report No.: 7415737-001 REV A1.

Danaci, D., Bui, M., Petit, C., Dowell Mac, N., 2021. En route to zero emissions for power and industry with amine-based post-combustion capture. Environ. Sci. Technol. 55 (15) https://doi.org/10.1021/acs.est.0c07261.

Direct Air Capture. Paris: International Energy Agency; 2022.

Donald, R.W., 2007. Rules of Thumb in Engineering Practice. Wiley VCH.

Elliott, W., Benzb, A., Gibbins, J., Michailos, S., 2021. An open-access, detailed description of post-combustion CO2 capture plant. In: Proceedings of the 15th International Conference on Greenhouse Gas Control Technologies. Abu Dhabi.

Elliott, W.R. Front-end engineering design (FEED) study for a carbon capture plant retrofit to a natural gas-fired gas turbine combined cycle power plant (2 2 1 Duct-Fired 758-MWe facility with F Class Turbines). https://www.energy.gov/: Bechtel; 2022 March 2022.

Evaluating the Costs of Retrofitting, 2017. CO2 Captured in an Integrated Oil Refinery: Technical Design Basis and Economic Assumptions. IEA Greenhouse Gas R&D Programme. Report No.: 2017-TR5.

Feron, P., Cousins, A., Jiang, K., Zhai, R., Hla, S.S., Thiruvenkatachari, R., Burnard, K., 2019. Towards zero emissions from fossil fuel power stations. Int. J. Greenhouse Gas Control 87, 188–202. https://doi.org/10.1016/j.ijggc.2019.05.018.

Fossil fuel price assumptions [Internet] https://www.gov.uk/: department for Business, Energy and Industrial Strategy; 2022 [Available from: https://www.gov.uk/government/collections/fossil-fuel-price-assumptions.

Fuss, S., F Lamb, W., Callaghan, M.W., Hilaire, J., Creutzig, F., Amann, T., et al., 2018. Negative emissions - Part 2: costs, potentials and side effects. Environ. Res. Lett. 13 (6) https://doi.org/10.1088/1748-9326/aabf9f.

Gao, T., Selinger, J.L., Rochelle, G.T., 2019. Demonstration of 99% CO2 removal from coal flue gas by amine scrubbing. Int. J. Greenhouse Gas Control 83, 236–244. https://doi.org/10.1016/j.ijggc.2019.02.013.

Gibbins, J., Lucquiaud, M., 2021. Post-combustion Carbon Dioxide capture: Best Available Techniques (BAT). UK Goverment. https://www.gov.uk. Contract No.: Guidance and support.

Gibbions, J..Lucquiaud, M. BAT Review for New-Build and Retrofit Post-Combustion Carbon Dioxide Capture Using Amine-Based Technologies for Power and CHP Plants Fuelled by Gas and Biomass as an Emerging Technology under the IED for the UK. https://ukccsrc.ac.uk: UKCCSRC; 2021 July 2021.

gPROMS products: gPROMS PROCESSBUILDER [Internet] 2021 [Available from: https://psenterprise.com/products/gproms/processbuilder.

Heap, R., Jackson, N., Bance, P., Gammer, D., 2016. Potential Role of Hydrogen in the UK Energy System. Energy Research Partnership.

Herraiz Palomino, L., Su, D., Muslemani, H., Struthers, I., Thomson, C., Chalmers, H., Lucquiaud, M., 2022. A preliminary assessment of negative CO2 emissions in the European waste sector. In: Proceedings of the 2nd International Conference on Negative CO2 Emissions. Göteborg, Sweden.

Hirata, T., Tsujiuchi, T., Kamijo, T., Kishimoto, S., Inui, M., Kawasaki, S., et al., 2019. Near-zero emission thermal power plant using advanced KM CDR processtm. Int. J. Greenhouse Gas Control 92, 102847.

Howarth, R.W, Jacobson, M.Z., 2021. How green is blue hydrogen? Energy Sci. Eng. 9 (10), 1676–1687. https://doi.org/10.1002/ese3.956.

Hydrogen production costs, 2021. Department for business. Energy Ind. Strat. https://www.gov.uk/.

Ismail Shah, M., Silva, E., Gjernes, E., Åsen Ingvar, K., 2021. Cost reduction study for MEA based CCGT post-combustion CO2 capture at technology center Mongstad. 15th Greenhouse Gas Control Technol. Conf. 15–18. Abu dhabi: SSRN.

Joffe, D., Livermorey, S., Hemsley, M. Hydrogen in a low-carbon economy. Committee on Climate Change; 2018 22 November 2018.

Léonard, G., Crosset, C., Toye, D., Heyen, G., 2015. Influence of process operating conditions on solvent thermal and oxidative degradation in post-combustion CO2 capture. Comput. Chem. Eng. 83, 121–130. https://doi.org/10.1016/j.compchemeng.2015.05.003.

Masson-Delmotte, V., Pirani, A., Chen, Y., Matthews, J.B.R., Yelekçi, O., Lonnoy, E., et al., 2021. Climate Change 2021: The Physical Science Basis. Cambridge University Press: IPCC.

Mcqueen, N., Vaz Gomes, K., Mccormick, C., Blumanthal, K., Pisciotta, M., Wilcox, J., 2021. A review of direct air capture (DAC): scaling up commercial technologies and innovating for the future. Prog. Energy 3 (3). https://doi.org/10.1088/2516-1083/abf1ce.

Michailos, S..Gibbins, J. A modelling study of post-combustion capture plant process conditions to facilitate 95–99% Co2 capture levels from gas turbine flue gases frontiers in energy research. 2022;10. 10.3389/fenrg.2022.866838.

Morgan, J.C., Chinen, A.C., Anderson-Cook, C., Tong, C., Carroll, J., Saha, C., et al., 2020. Development of a framework for sequential Bayesian design of experiments: application to a pilot-scale solvent-based CO2 capture process. Appl. Energy 262. https://doi.org/10.1016/j.apenergy.2020.114533.

Mullen, D., Lucquiaud, M., 2022. Zero carbon electricity with CCS: a techno-economic analysis of combined cycle gas turbines. In: Proceedings of the 14th International Conference on Applied Energy (ICAE2022). Germany. Bochum.

National Carbon Capture Center [Internet] 2021 [Available from: https://www.nationalcarboncapturecenter.com/.

Negative Emissions, 2019. Technologies and Reliable Sequestration: A Research Agenda. The National Academies Press: National Academies of Sciences, Engineering, and Medicine, Washington, DC.

Nessi;, E., Papadopoulos;, A.I., Seferlis, P., 2021. A review of research facilities, pilot and commercial plants for solvent-based post-combustion CO2 capture: packed bed, phase-change and rotating processes. Int. J. Greenhouse Gas Control 111. https://doi.org/10.1016/j.ijggc.2021.103474.

Nexant, 2016. Pre-Feasibility Study for Establishing a Carbon Capture Pilot Plant in Mexico. World Bank. https://www.gob.mx.

Ofgem. Gas Prices: forward Delivery Contracts - Weekly Average (GB) ofgem.gov.uk 2022 [Available from: https://www.ofgem.gov.uk/energy-data-and-research/data-portal/wholesale-market-indicators.

Rates, E., 2023. US Dollar to British Pound Spot Exchange Rates for 2020 [Internet]. Exchange Rates. Available from: https://www.exchangerates.org.uk/USD-GBP-spot-exchange-rates-history-2020.html.

Rehl, T., Müller, J., 2013. CO2 abatement costs of greenhouse gas (GHG) mitigation by different biogas conversion pathways. J. Environ. Manage. 114, 13–25. https://doi.org/10.1016/j.jenvman.2012.10.049.

Review of next generation, 2022. carbon capture technology for industrial, waste and power sectors. www.GOV.UK. Dep. Bus. Energy Ind. Strat.

Romano, M.C., Antonini, C., Bardow, A., Bertsch, V., Brandon, N.P., Brouwer, J., et al., 2022. Comment on "How green is blue hydrogen? Energy Sci. Eng. 10 (7), 1944–1954. https://doi.org/10.1002/ese3.1126.

Sinnott, R., Towler, G., 2020. Chemical Engineering Design. Elsevier.

Smith, M.A., Cain, M., Allen, M.R., 2021. Further improvement of warming-equivalent emissions calculation. Clim. Atmos. Sci. 4 https://doi.org/10.1038/s41612-021-00169-8.

Soltani, S.M., Fennell, P.S., Mac Dowell, N., 2017. A parametric study of CO2 capture from gas-fired power plants using monoethanolamine (MEA). Int. J. Greenhouse Gas Control 63, 321–328. https://doi.org/10.1016/j.ijggc.2017.06.001.

Spath, P.L., Mann, M.K., 2001. Life Cycle Assessment of Hydrogen Production Via Natural Gas Steam Reforming. National Renewable Energy Laboratory. Report No.: NREL/TP-570-27637.

Stark, C., Mike, T..Joffe, D. The sixth carbon budget: the UK's path to net zero. Committee on Climate Change; 2020 9 December 2020.

THE CHEMICAL ENGINEERING, 2022. PLANT COST INDEX [Internet] Chemengonline. Com. Chemical Engineering. Available from: https://www.chemengonline.com/pci-home.

UK Low carbon hydrogen standard. www.gov.uk: Crown copyright; 2022.

UK Hydrogen, 2021. Strategy. www.gov.uk: department for business. Energy Ind. Strat.