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Pathways to low-cost clean hydrogen production with gas switching reforming

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HIGHLIGHTS

- Hydrogen plant with gas switching reforming is optimised for efficiency and cost.
- GSR-H2 can achieve negative specific primary energy consumption for CO2 avoided.
- GSR-H2 produces 4.5% cheaper H2 with 96% CO2 capture at 15 $ per ton CO2 avoided.
- GSR-H2 technology is robust to almost any future energy market scenario.
- Oxygen carrier stability is critical for techno-economic performance of GSR-H2 plant.

ABSTRACT

Gas switching reforming (GSR) is a promising technology for natural gas reforming with inherent CO2 capture. Like conventional steam methane reforming (SMR), GSR can be integrated with water-gas shift and pressure swing adsorption units for pure hydrogen production. The resulting GSR-H2 process concept was techno-economically assessed in this study. Results showed that GSR-H2 can achieve 96% CO2 capture at a CO2 avoidance cost of 15 $/ton (including CO2 transport and storage). Most components of the GSR-H2 process are proven technologies, but long-term oxygen carrier stability presents an important technical uncertainty that can adversely affect competitiveness when the material lifetime drops below one year. Relative to the SMR benchmark, GSR-H2 replaces some fuel consumption with electricity consumption, making it more suitable to regions with higher natural gas prices and lower electricity prices. Some minor alterations to the process configuration can adjust the balance between fuel and electricity consumption to match local market conditions. The most attractive commercialization pathway for the GSR-H2 technology is initial construction without CO2 capture, followed by simple retrofitting for CO2 capture when CO2 taxes rise, and CO2 transport and storage infrastructure becomes available. These features make the GSR-H2 technology robust to almost any future energy market scenario.

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Anthropogenic CO₂ emissions have been the dominant cause of observed global warming from the middle of 20th century [1]. The International Energy Agency (IEA) presented different pathways and their contributions to mitigate these emissions, of which, fuel switching and carbon capture and storage (CCS) accounts for reduction of 5% and 14% (respectively) of the cumulative global CO₂ emissions by 2060 in the defined 2° scenario [2]. When global temperatures must be restricted below 1.5 °C [3], the Intergovernmental Panel on Climate Change (IPCC) presented scenarios with more emphasis on hydrogen and carbon capture utilization and/or storage (CCUS) as pathways to mitigate CO₂ emissions.

Hydrogen is attracting increasing attention as a carbon-free energy carrier in industry, transport and power sector, exemplified by a recent IEA special report [4]. In this report, natural gas reforming with conventional CCS remains economically more attractive than electrolytic hydrogen production in most regions over coming decades. This assessment does not account for a next generation of natural gas reforming technologies with integrated CO₂ capture that have the potential to achieve similar costs to reference plants without CO₂ capture (e.g. membrane-assisted reforming technologies [5]). Another such technology is assessed in this study: gas switching reforming (GSR).

The demand for hydrogen has been on a rise since 1975 as seen in Fig. 1, mainly in the ammonia and oil refining sector. It is predicted to further increase with favorable policy directives for hydrogen deployment, especially in the transport sector [4]. 76% of the current hydrogen in the world is produced from natural gas (NG) reforming that results in 10 ton of CO₂ emitted per ton of H₂ produced [4]. The IEA predicts that NG will overtake coal in 2030 to become the second largest fuel (after oil) in global energy mix, of which the industrial consumers will contribute to a 45% increase in worldwide gas use [6]. In this scenario, hydrogen production from NG with CCUS, which is the focus of this study, provides an attractive clean energy alternative. Hydrogen also provides an opportunity to use existing gas infrastructure for new clean hydrogen supplies [4], especially related to transport and storage of it.

A detailed review on different hydrogen production methods integrated with CO₂ capture was presented by Voldsund et al. [7]. In conventional hydrogen production process, NG is first converted to syngas via steam methane reforming (SMR) in fired tubular reformer (FTR) or auto-thermal reformer (ATR). FTR uses external firing to provide heat for the reaction whereas in ATR, a fraction of the fuel is combusted internally to provide heat. Syngas is converted to CO₂ and H₂ in subsequent water-gas shift (WGS) reactors. Pure hydrogen is recovered in a pressure swing adsorption (PSA) step and the off-gas is combusted either to provide heat for reforming reaction or in a combined heat and power (CHP) unit. SMR with FTR is currently the dominant technology to produce large scale pure hydrogen from NG due to its favorable economics [4]. In the SMR process, CO₂ can be captured from the upstream or downstream of H₂ recovery step (reducing CO₂ emissions by 60%) or the exhaust gases from FTR burners (reducing CO₂ emissions by 90%). Each of these
gas streams has different conditions with respect to partial pressure of CO₂ and the total mass flow of CO₂. Soltani et al. [8] reported that capturing CO₂ at the upstream of the H₂ recovery step is more favorable for the design conditions (Steam/Carbon mole ratio of 2.5 - 3 and reformer temperature of 890 °C) in the conventional SMR plant. However, when CO₂ is captured from the exhaust gases of FTR to obtain ~80% CO₂ avoidance, the equivalent hydrogen production efficiency of the SMR process drops by 14 %-points when compared to the reference plant and thereby increasing the cost of hydrogen by 30% [5]. The penalty is due to capturing CO₂ from dilute flue gases that contain mostly N₂. Similar penalty is also reported for methane-cracking technology studied by Abanades et al. [9] to produce clean hydrogen, but in this process the main challenge is with handling of carbon deposits. This penalty is minimized in ATR based H₂ production process by having an air separation unit (ASU) that feeds in pure oxygen for reforming and boiler sections [10]. The flue gas contains only CO₂ and H₂O, from which, H₂O is condensed leaving pure CO₂ stream for transport and storage. Although the penalty in capturing CO₂ is only ~2%-points in the ATR based process, it requires an additional ASU that has high capital costs alongside leaving the N₂ stream that is not used in the process. Therefore, ATR is more suited for ammonia production that does need separation of N₂ in ASU [11]. The hydrogen production costs from these two technologies are sensitive to the NG price and the CO₂ tax (when comparing with options with CO₂ capture) [12].

Membrane and sorption enhanced reactors are alternatives for integrated syngas production and gas separation [7]. These methods can be applied to either SMR or WGS steps. WGS and SMR membrane reactors separate the H₂ whereas sorption enhanced WGS or SMR reactors separate CO₂. The membrane reactors produce pure H₂ leaving an impure CO₂ stream, and the sorption enhanced reactors produce high quality CO₂ stream for transport and storage but a low purity H₂ stream. In addition, membranes are cost intensive and the sorption enhanced system requires highly active solid sorbents for higher hydrogen production efficiencies [13]. Pressurized sorption enhanced systems may have lower costs due to compact systems and lower work required for H₂ and CO₂ compression, but the equivalent hydrogen efficiency is lower. A variant of sorption enhanced hydrogen production system is the Ca-Cu looping process that consists of three stages, the sorption enhanced reforming, Cu oxidation with air and CuO reduction alongside CaCO₃ calcination. The Ca-Cu looping process for hydrogen production exhibits specific primary energy consumption for CO₂ avoided (SPECCA) of 1.07 - 1.54 MJ/kg-CO₂ depending on the pressure inside the reactor [14]. It can capture ~96% CO₂ at 31 €/ton cost of CO₂ avoided. However, the cost of hydrogen produced using Ca-Cu looping process is 16% higher than in SMR plant without capture [14].

Chemical looping reforming (CLR) systems show higher potential to produce syngas and inherently separate air [15]. A schematic of a typical CLR system is shown in Fig. 2a. NG and steam are reformed in the fuel reactor of the CLR to produce syngas in the presence of an oxidised metallic oxygen carrier. The oxygen carrier is reduced and sent to the oxidation reactor where it is oxidised by the air stream, leaving a depleted air stream that mainly contains N₂. In addition, Kang et al. [16] analysed a three-reactor chemical looping reforming system comprising of air, fuel and steam reactors. The oxidised metal oxide (oxygen carrier) from the air reactor is partially reduced in the fuel reactor with methane forming CO₂ and H₂O, and the remaining reduction of the metal oxide happens in the steam reactor where steam reduces the oxygen carrier to form H₂. One of the main practical challenges with these CLR systems is their scale-up for pressurized interconnected operation, which is needed for higher process efficiency [17]. When CLR is integrated in a process for gas-fired power production, it needs to be operated between 18 and 20 bar [18], whereas hydrogen production processes require even higher pressures [17].

To address the challenge with respect to circulation of oxygen carrier between inter-connected reactors in chemical looping, several other reactor configurations including packed bed chemical looping [19,20], rotating bed reactor [21] and gas switching reactor [22,23] have been developed and presented in literature for combustion or reforming of fuel. The present study focuses on the application of gas switching to NG reforming to produce hydrogen. A schematic of the gas switching reforming (GSR) system is shown in Fig. 2b. The oxygen carrier (Ni-NiO system in this study) is kept inside the
bubbling fluidized bed reactor, and the gas streams alternate during different oxidation, reduction and reforming stages. The air stream oxidises the oxygen carrier, producing an oxygen depleted air stream \((\text{N}_2 \text{ stream})\) during the oxidation step. Subsequently, the oxygen carrier is reduced by the off gases from the pressure swing adsorption (PSA) unit during the reduction step. The reduced oxygen carrier then acts as a catalyst for the steam-methane reforming in the third step of the GSR process. There is no circulation of oxygen carrier between reactors that eliminates the challenges with loop seals and cyclones. GSR also allows for easy integration of the PSA unit to recover high purity hydrogen and use the off-gases in the reduction step. However, GSR requires high temperature inlet and outlet valves to switch gases between different stages. In addition, GSR needs a cluster of reactors to provide steady product output from the process [24].

The GSR reactor was experimentally demonstrated by Wassie et al. [23], where the effect of reactor temperature and cycle times (for oxidation, reduction and reforming steps) on the conversion of methane was reported. A 1D phenomenological model for GSR was developed to understand the flow, temperature and conversion profiles inside the reactor [25] and compared against the conventional CLR process. GSR has lower fuel conversion but produces syngas with higher heating value. Following this, the potential of GSR process integrated with combined cycle gas-fired power production and CO2 capture for pure hydrogen production in a process designated as GSR-H2 [17], and its technical performance was compared against the conventional SMR process. In case of no CO2 capture, GSR-H2 has nearly 3%-points higher equivalent hydrogen production efficiency when compared to SMR. In addition, GSR-H2 with more than 96% CO2 capture faces an efficiency penalty of only 0.3%-points when compared to SMR plant without capture. However, GSR-H2 process consumes more electricity and hence, the source of electricity is critical for the performance of the GSR-H2 process. This will not only affect the efficiency of the process but will also affect the cost of hydrogen. In future energy scenarios, the cost of NG and CO2 tax are expected to increase that will also have an impact on the cost of hydrogen produced [28]. A first of its kind techno-economic analysis revealed that GSR-H2 has the potential to produce hydrogen with similar costs to that from SMR plant without capture [29]. However, there is still a gap in literature with respect to the detailed techno-economic performance of the GSR-H2 process and its performance in future energy scenarios (for example: availability of cheap renewable electricity, CO2 tax uncertainty etc). This paper improves upon the previous work on GSR-H2 process [17,29] and fills the gap in literature by carrying out process efficiency improvement studies of the GSR-H2 process through design changes, detailed techno-economic analysis and presenting scenarios for deployment of this technology. Equivalent hydrogen production efficiency, CO2 avoidance, cost of CO2 avoidance and levelised cost of hydrogen are the

**Fig. 2** – Schematic of a) Chemical looping reforming b) Gas switching reforming.
key performance indicators for techno-economic assessment of the GSR-H2 technology. The results have been compared against the reference SMR plant.

Process description of reference plant, base case GSR-H2 and improved GSR-H2 process

Reference steam methane reforming (SMR) plant

The conventional SMR plant for hydrogen production has been described previously in literature [5,13]. NG is desulphurized, mixed with steam and pre-heated for pre-reforming step to convert higher hydrocarbons into CH₄. The steam and CH₄ mixture (steam to carbon ratio of 2.70) is sent for reforming inside a catalytic fired tubular reformer (FTR), where more than 80% of CH₄ is converted to form syngas. CO and H₂O in the syngas are converted to CO₂ and H₂ in the WGS reactor before H₂ is separated in the PSA. Pure H₂ recovered from the PSA is compressed for transport and storage, whereas the PSA off-gas is combusted in the FTR burners to provide necessary heat for the reforming step. There is a lot of heat available in the process which is recovered to produce superheated high-pressure steam. A fraction of the steam is used in reforming and the remainder is expanded in steam turbines to generate electricity. The SMR plant simulation has been reproduced in this study, using the modelling assumptions in Nazir et al. [17], to maintain consistency in results obtained.

Base case GSR-H2 process

The schematic of the base case GSR-H2 is shown in Fig. 3. The pre-reforming section is similar to the SMR plant with a slightly lower steam to carbon (S/C) ratio of 2.66 at the inlet of the reforming step of the GSR. The outlet from the pre-reformer is pre-heated to 825°C and fed to the GSR. GSR is designed at 32.7 bar, similar to the pressure conditions in the FTR in SMR plant. Syngas from the reforming step of GSR is cooled to 302°C and sent to WGS step to convert nearly 77%-mol of CO into CO₂ and H₂O into H₂. The syngas from the WGS reactor is cooled to 25°C to condense all the water from the stream to avoid degradation of the PSA adsorbents. 87.9% of H₂ is recovered in the PSA and compressed in three stages until 150 bar and 25°C for transport and storage [5]. The off-gas from the PSA is compressed and pre-heated to 1000°C before being fed to the GSR for the reduction step. The reduction step product gas contains only CO₂ and H₂O. H₂O is condensed and the CO₂ stream is compressed until 110 bar and 25°C for transport and storage. Compressed air oxidises the oxygen carrier during the oxidation step of the GSR leaving the depleted air stream containing mainly N₂. The N₂ stream from the GSR is cooled and expanded in a gas turbine to generate electricity. There is a lot of heat that can be recovered from the process to pre-heat the process streams as well as prepare steam for reforming. The heat recovered in cooling the syngas from the GSR is used to pre-heat the NG and steam mixture at the inlet of the GSR, pre-reformer inlet and NG before the desulphurization step. The WGS product and the N₂ stream from the oxidation step of GSR are cooled down to generate steam for reforming. The outlet from the reduction step of the GSR that mainly contains CO₂ and H₂O, preheats the PSA off-gas that is fed to the GSR. The remaining heat in the reduction stage outlet is also used to generate steam for reforming.

The GSR-H2 process has higher electrical consumption when compared to the SMR plant. This is due to the work required for compressing air and CO₂ streams in addition to the H₂ stream as in SMR plant. Therefore, the source of electricity affects the overall process performance of GSR-H2. Efficient electricity source like NGCC results in higher equivalent hydrogen production efficiency of GSR-H2, whereas using a standalone NG boiler to fulfil electricity demand might result in an additional 2%-point efficiency penalty [17]. In addition to the efficiency penalty, the CO₂ avoidance also reduces. The sensitivity of levelised cost of hydrogen from GSR-H2 with respect to the source and cost of electricity is discussed later in this paper.

Improvements in design of GSR-H2 process

Four different designs of the GSR-H2 process are proposed to improve its techno-economic performance. In case 1, steel rods are inserted inside the GSR to increase the overall heat capacity of the GSR reactor system. This added thermal mass minimizes the temperature drop during the endothermic reforming step, achieving a higher average reforming temperature to lower the S/C ratio required for high fuel conversion. In case 2, a two-phase evaporator [24] is considered to produce steam for reforming using the steam condensation enthalpy in the CO₂ stream from the GSR reduction step that would otherwise be rejected. Case 3 is a combination of the above two designs, and case 4 presents an alternative heat integration strategy for the N₂ stream from the GSR oxidation step to increase H₂ production at the cost of increased power consumption. These designs are discussed briefly below.

Case 1: GSR with additional thermal mass

The GSR reactor is assumed to be inserted with steel rods to double the heat capacity in the reactor, which requires about 25% of the reactor volume to be occupied with steel rods [17]. This modification results in overall higher temperatures in different stages of GSR. Therefore, required conversion of methane is achieved at lower S/C ratios [17]. The main difference between the base case GSR-H2 process (as shown in Fig. 3) and this case is that the steam required for reforming is generated completely from cooling the WGS product (Boiler 1 in Fig. 3) and the GSR reduction stage outlet streams (Boiler 2 in Fig. 3). Therefore, the N₂ stream from the GSR oxidation stage is directly expanded in the turbine to generate a larger quantity of electricity. The remaining heat in the N₂ stream after expansion is recovered to generate saturated 6 bar steam for export.

Case 2: GSR-H2 process with two-phase evaporator to generate steam

In this case, the GSR reactor system is similar to the base case, without any additional thermal mass. A two-phase evaporator is used to generate steam for reforming while recovering heat from the GSR reduction stage outlet. The base case GSR-H2
The NG needs to be modified to include this design change. NG is pre-heated for desulphurization with the desulphurized NG stream and the N₂ stream from the GSR oxidation step. The desulphurized NG and pressurized water stream is sent to the two-phase evaporator with the GSR reduction step outlet stream on the hot side of the evaporator. The water evaporates at low temperatures due to the presence of non-condensable fuel gases that reduce the steam partial pressure in the stream, allowing the steam condensation enthalpy in the CO₂ stream to be efficiently used for steam generation. The NG-steam mixture is then mixed with additional remaining steam, which is generated by recovering heat from the WGS product and N₂ stream, before being sent for pre-heating followed by pre-reforming step. Considering these modifications, the GSR-H₂ process in case 2 would not have Economiser 2 and 3 and Boiler 2 as shown in Fig. 3, but instead have a two-phase evaporator. The Hex 1 in Fig. 3 is also replaced with two heat exchangers, where NG exchanges heat with desulphurized NG and N₂ stream.

**Case 3: GSR-H₂ process with additional thermal mass and two-phase evaporator**

The GSR-H₂ in case 3 is a combination of cases 1 and 2. Desulphurized NG and water mixture is passed through the two-phase evaporator that recovers steam condensation enthalpy from the GSR reduction step outlet. The GSR reduction step also operates at a higher average temperature, allowing for a lower S/C ratio (lower heat requirement for steam generation). The hot N₂ stream from the oxidation step of GSR is expanded in the gas turbine to generate electricity. Remaining heat in the N₂ stream after expansion is recovered to generate saturated 6 bar steam for export.

**Case 4: GSR-H₂ process as in case 3 in addition to pre-heated air for oxidation step of GSR**

Case 3 is modified by including an air pre-heating step before the GSR oxidation stage so that the heat available in the process is directed towards higher conversion of methane in the GSR instead of high power production from expanding the N₂ stream at a high temperature. This configuration also eliminates the need to export a substantial amount of heat as low-value 6-bar steam. The design in case 4 is defined as advanced GSR-H₂ process. The schematic of the advanced GSR-H₂ process is shown in Fig. 4.

### Methodology and assumptions

This section is presented in three parts. The first two parts describe the modelling methodology and assumptions in developing the 0D model for the GSR and the process model for the reference SMR plant and GSR-H₂ process. The third part describes the methodology and assumptions used to carry out the techno-economic assessment of the GSR-H₂ process.

**0D model for the GSR**

The transient gas switching reforming process is simulated using a 0D model coded in Matlab R2018b. The model solves the mass and energy balances of the system, assuming continuous stirred tank reactor (CSTR) behaviour of the reactor and that chemical- and thermal equilibrium are reached. These assumptions are reasonable considering the excellent mixing behaviour of fluidized bed reactors, the fast
reaction kinetics of the NiO oxygen carrier that is considered and the large geometries of industrial-scale fluidized beds [17]. This reactor model has been used in several previous studies [17,24,26], therefore the detailed model description is not repeated here. The interested reader is referred to these earlier studies for a complete description of the equations that are solved and the coupling between the reactor and process models.

The reactions occurring inside the GSR reactor are listed below. Eqs. (1)–(3) occur during the reduction step, Eq. (4) in the oxidation step, and Eqs. (5)–(7) in the reforming step.

\[
\begin{align*}
\text{CH}_4 + 4\text{NiO} & \rightarrow 4\text{Ni} + \text{CO}_2 + 2\text{H}_2\text{O} \quad (1) \\
\text{H}_2 + \text{NiO} & \rightarrow \text{Ni} + \text{H}_2\text{O} \quad (2) \\
\text{CO} + \text{NiO} & \rightarrow \text{Ni} + \text{CO}_2 \quad (3) \\
\text{O}_2 + 2\text{Ni} & \rightarrow 2\text{NiO} \quad (4) \\
\text{CH}_4 + \text{H}_2\text{O} & \rightarrow \text{CO} + 3\text{H}_2 \quad (5) \\
\text{CO} + \text{H}_2\text{O} & \rightarrow \text{CO}_2 + \text{H}_2 \quad (6) \\
\text{CH}_4 + 2\text{H}_2\text{O} & \rightarrow \text{CO}_2 + 4\text{H}_2 \quad (7)
\end{align*}
\]

The GSR reactors are operated similar to a previous study [17]. Firstly, the air flow rate is controlled to maintain a maximum reactor temperature of 1100 °C to prevent degradation of the oxygen carrier. The percentage oxygen carrier utilization is set equal to the reactor pressure in bars, which is kept constant at 32.7 bar for all cases considered here. The switching of the outlet valves between steps is slightly delayed compared to the switching of the inlet valves, since it has been shown that such a strategy reduces the undesired mixing between different reactor steps [30]. The ratio of the stage times of the reduction, reforming and oxidation steps (and therefore also the ratio of the number of reactors operating in each step) is specified as 1:3:1. This ratio results in similar outlet flow rates during each of the steps, which is required to produce a sufficiently steady output in combination with the delayed outlet switching. Finally, a GSR reactor cluster of 10 reactors (diameter = 1.23 m, height = 3.69 m) is considered. The chosen reactor diameter will operate the reactor in the bubbling fluidization regime with fluidization velocities of around 0.4 m/s when a conventional particle size of 150 μm is used.

Fig. 5a shows a cycle of the GSR reactor. In the reduction step (0–1 on the x-axis), the PSA off-gas is combusted to CO2 and H2O, reducing the oxygen carrier. Subsequently, in the reforming step (1–4 on the x-axis), the methane is reformed to syngas. Since the reforming reactions are highly endothermic, the reactor temperature falls rapidly in the reforming step. Finally, in the oxidation step (4–5 on the x-axis), the oxygen carrier is oxidised by air, rapidly heating the reactor again. The undesired mixing between the steps can also be seen in Fig. 5. The primary consequences of the mixing are that 1) CO2 from the reforming step mixes into the oxidation step outlet stream, reducing the CO2 capture efficiency, and 2) N2 from the oxidation step mixes into the reduction step outlet stream, reducing the CO2 purity.

Fig. 5 also compares the two most efficient cases (namely case 3 and 4 in section Improvements in design of GSR-H2 process) considered in the present study: those with added thermal mass in the reactor and with a two-phase heat exchanger in the process configuration. The difference
between the two panels is that for the case in panel b) the air stream is pre-heated before entering the reactor. This slightly increases the temperature fluctuation over the cycles, since less energy is required to heat the incoming air, causing a greater temperature increase in the oxidation step for the same oxygen carrier utilization.

It can be noted that a previous study [17] already showed that adding additional thermal mass to the reactor leads to a large reduction in the temperature variation across the cycle. This results in a higher average reactor temperature in the reforming step, which leads to a higher methane conversion. Furthermore, it can be noted that adding the two-phase heat exchanger to the process configuration had a negligible effect on the reactor cycle, therefore the results for cases without the two-phase heat exchanger are not shown here.

**Process model for SMR and GSR-H2**

The SMR and GSR-H2 process are simulated and analysed using Aspen Hysys V8.6 [31]. Peng-Robinson thermodynamic model is used to estimate the properties of components and mixtures at equilibrium. The components in the process including heat exchangers, boilers, reactors, compressors and expanders are modelled as per the assumptions used in Nazir et al. [17]. The PSA has been modelled as a black box and the recovery of 99.999% pure H$_2$ is estimated using Eq. (8) [17], where $P_1$ and $P_2$ are the pressure of the PSA inlet and off-gas stream respectively. One natural draft cooling tower is considered for each process, in which the cooling water temperature rise is assumed to be 12°C.

$$
H_2 \text{ recovery in PSA (\%)} = 100 - \frac{100}{0.2521 \left( \frac{P_1}{P_2} \right) + 1.2706}
$$

Methodology for techno-economic analysis

Techno-economic analysis deals with assessment of potential of a technology in early stages of its development. The main key performance indicators to assess and compare hydrogen production processes are hydrogen production efficiency, equivalent hydrogen production efficiency, CO$_2$ capture and avoidance, specific primary energy consumption for CO$_2$ avoided (SPECCA), and economic indicators of levelised cost of hydrogen (LCOH) and cost of CO$_2$ avoidance (COCA). The method to estimate these key performance indicators is described below and is divided in two sections: process analysis methodology and economic analysis methodology. The basis for analysis is 10 TPH NG input to the process.

Process analysis methodology

Firstly, equivalent NG ($m_{eq,NG}$) is calculated using Eq. (9) that accounts not only for the NG consumed in the process but also for steam exports from the process and the net electrical work.

$$
Q_{th} = \frac{m_{steam\text{ export}} \times (h_{steam\text{ export}} - h_{liq\text{ at}6\text{ bar}})}{0.583} \quad (10)
$$

where $h_{steam\text{ export}}$ and $h_{liq\text{ at}6\text{ bar}}$ are the enthalpies of the steam export and the liquid at 6 bar respectively.

The hydrogen production efficiency ($\eta_{h}$) and the equivalent hydrogen production efficiency ($\eta_{eq,h}$) are calculated using Eq. (11) and Eq. (12).

$$
\eta_h = \frac{Q_h}{W_d}
$$

$$
\eta_{eq,h} = \frac{Q_h}{W_d + \Delta W_d}
$$

**Fig. 5** – The temperature and composition as a function of the number of reduction step lengths for a full cycle of the GSR for a) without and b) with air preheating. The reduction step lengths are 101 s and 79 s respectively. Both cases consider the reactor with added thermal mass and the plant configuration with a two-phase heat exchanger.
\[ \eta_{3} = \frac{100% \times m_{H2} \times LHV_{H2}}{m_{NG} \times LHV_{NG}} \]  
(11)

\[ \eta_{eq,3} = \frac{100% \times m_{H2} \times LHV_{H2}}{m_{eq,NG} \times LHV_{NG}} \]  
(12)

The amount of CO₂ captured (CC) and CO₂ avoided (CA) is calculated using Eq. (13) and Eq. (14). The CO₂ emission intensities of NG combustion \(E_{NG} = 56.8 \text{ gCO₂/MJ LHV}\), steam generation \(E_{\text{steam}} = 63.3 \text{ gCO₂/MJ}\) and electrical work consumed \(E_{\text{elec}} = 97.7 \text{ gCO₂/MJ}\) are used to calculate the CO₂ avoided from the process. The values of \(E_{\text{NG}}\) and \(E_{\text{elec}}\) will change if the factor for energy conversion of NG into steam (0.9 as in Eq. (9)) and electricity (0.583 as in Eq. (9)) is changed.

\[ CC = \frac{100\% \times \text{mass of CO}_2 \text{captured}}{m_{NG} \times LHV_{NG} - E_{NG} - Q_{\text{th}} - W_{\text{el}} - E_{\text{elec}}} \]  
(13)

\[ CA = \frac{100\% \times \text{mass of CO}_2 \text{captured}}{m_{H2} \times LHV_{H2} - Q_{\text{th}} - W_{\text{el}} - E_{\text{elec}}} \]  
(14)

The CO₂ emission intensity of the process and its equivalent in terms of \(\text{gCO}_2/\text{MJ}\) is calculated using Eq. (15) and Eq. (16), whereas the SPECCA is calculated using Eq. (17).

\[ E_{\text{CO}_2} = \frac{\text{mass of CO}_2 \text{ emitted}}{m_{H2} \times LHV_{H2}} \]  
(15)

\[ E_{\text{eq,CO}_2} = \frac{\text{mass of CO}_2 \text{ emitted} - Q_{\text{th}} - W_{\text{el}} - E_{\text{elec}}}{m_{H2} \times LHV_{H2}} \]  
(16)

\[ \text{SPECCA} = 1000 \times \frac{1}{\frac{E_{\text{eq,CO}_2}}{E_{\text{eq,CO}_2 \text{ ref}}} - 1} \]  
(17)

**Economic analysis methodology**

The economic analysis for the GSR-H₂ process and its comparison with the conventional SMR technology is carried out in accordance with the methodology proposed by the Global CCS Institute [33]. LCOH is calculated using Eq. (18).

\[ \text{LCOH} = \frac{(TCR) \times FCF + FOM \times (FC) \times (HR)}{(m_{H2}) \times (CF^*8766)} + VOM \]  
(18)

The definition of the terms used in Eq. (18) is presented in Table 1. The fixed charge factor (FCF) is calculated using Eq. (19) that converts the total capital value into uniform annual amounts. The FCF is a function of discount rate and lifetime of the plant. In this study a discount rate of 10% and lifetime of 30 years have been assumed.

\[ FCF = \frac{r(1+r)^T}{(1+r)^T - 1} \]  
(19)

The total capital requirement (TCR) for the process is estimated using the method described in Table 2 based on the bare erected cost (BEC) of the equipment defined as the sum of the installed cost of the equipment. The costs have been adjusted to the year 2019 costs using chemical engineering plant cost index (CEPCI) factors [34]. The installed costs for heat exchangers, boilers, air compressors and N₂-turbine is taken from the databases available in the economics tool in ASPEN Hysys V8.6 [31] and the PEACE component of the Thermoflow suite V26 [35]. The reference costs for desulphurization unit, pre-reformer, FTR, WGS reactor, PSA and hydrogen compressors are referred from Spallina et al. [5]. The installed costs for PSA off-gas compressor, CO₂ compression step and steam turbines is referred from Szima et al. [27]. GSR reactor costs are also estimated using the same methodology as presented in Szima et al. [27] using cost correlations from Turton et al. [36]. In the cases with added thermal mass, stainless steel rods were included in the reactor cost using a purchase price of 1725 $/ton, based on an online search. The purchase cost was tripled to estimate installed costs, which is similar to the practice employed in Turton et al. [36].

A process contingency of 30% is assumed for the GSR reactor since this reactor concept has been demonstrated on lab scale [23,37] and is being proposed for pilot scale demonstration. Process contingencies for all the other sections of the GSR-H₂ plant and the reference plant is 0% since the process technology is already in commercial use. The project contingency is assumed 10% [33] for both the processes since a complete process and engineering design can be easily developed during the project phase. Owner’s costs may include land and financing costs, inventory capital and start-up costs and accounts for

<p>| Table 1 – Definition of terms in Eq. (18) to calculate the levelised cost of hydrogen (LCOH). |</p>
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Definition</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>TCR</td>
<td>Total Capital Requirement in the base year of the analysis</td>
<td>$</td>
</tr>
<tr>
<td>FCF</td>
<td>Fixed Charge Factor as defined in Eq. 19</td>
<td>fraction</td>
</tr>
<tr>
<td>FOM</td>
<td>Fixed O&amp;M costs</td>
<td>$/year</td>
</tr>
<tr>
<td>mH2</td>
<td>Mass of hydrogen produced from the process</td>
<td>kg/hr</td>
</tr>
<tr>
<td>CF</td>
<td>Capacity Factor – availability of the plant</td>
<td>Fraction</td>
</tr>
<tr>
<td>VOM</td>
<td>Variable O&amp;M costs excluding the fuel costs</td>
<td>$/kg-H₂</td>
</tr>
<tr>
<td>HR</td>
<td>Net heat rate of the plant</td>
<td>MJ/kg-H₂</td>
</tr>
<tr>
<td>FC</td>
<td>Fuel Cost per unit of energy</td>
<td>$/MJ</td>
</tr>
<tr>
<td>r</td>
<td>Interest or discount rate</td>
<td>%</td>
</tr>
<tr>
<td>T</td>
<td>Economic lifetime of the plant relative to its base year</td>
<td>years</td>
</tr>
</tbody>
</table>

<p>| Table 2 – Methodology and assumptions to estimate the total capital requirement (TCR). |</p>
<table>
<thead>
<tr>
<th>Component</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bare Erected Cost (BEC)</td>
<td>Sum of installed cost of equipment 8% of BEC</td>
</tr>
<tr>
<td>Engineering Procurement Construction Costs (EPCC)</td>
<td>30% of BEC for GSR reactor; 0% for reference SMR plant and for remainder of the GSR-H₂ process</td>
</tr>
<tr>
<td>Process Contingency</td>
<td>10% of (BEC + EPCC + Process Contingency)</td>
</tr>
<tr>
<td>Project Contingency</td>
<td>Process Contingency + Project Contingency</td>
</tr>
<tr>
<td>Total Contingencies</td>
<td>Process Contingency + Project Contingency</td>
</tr>
<tr>
<td>Total Plant Costs (TPC)</td>
<td>BEC + EPCC + Total Contingencies</td>
</tr>
<tr>
<td>Owner’s Cost</td>
<td>20.2% of TPC [36]</td>
</tr>
<tr>
<td>Total Overnight Costs (TOC)</td>
<td>TPC + Owners Cost</td>
</tr>
<tr>
<td>Total Capital Requirement (TCR)</td>
<td>1.14*TOC [38]</td>
</tr>
</tbody>
</table>
20.2% of the total plant cost (TPC) in power plants as reported in the cost methodology adopted by DOE/NETL [38]. Similar owner’s cost is assumed in this study. The total overnight cost (TOC), which is the sum of TPC and owner’s cost, is multiplied by a factor of 1.14 to obtain the TCR. The factor 1.14 represents the increase in capital due to both escalation and interest during construction of an investor owned utility [38].

The assumptions used to estimate the fixed (FOM) and variable (VOM) operating and maintenance costs are listed in Table 3. To calculate the labour costs, 3 working shifts with 12 people in each are considered for the reference plant. Additional 3 people per shift are considered for the CO2 compression section in the GSR-H2 plant with CO2 capture. The NG price is assumed to be 6.9 $/GJ-LHV, but this is region and time specific, as is the electricity price. A sensitivity study for different NG and electricity price is presented in the results and discussion section. All the other assumptions listed in Table 3 are similar to previous studies [5,26] except that the euro to US dollar currency conversion rate is assumed to be 1.15 USD/EUR.

After calculating the LCOH of the processes, the cost of CO2 avoided (COCA) is calculated using Eq. (20). The difference between the LCOH of the GSR and reference plants is divided by difference in specific CO2 emissions in the plants to calculate the COCA. Although the LCOH will generally be reported including the CO2 tax shown in Table 3, the COCA reported in Table 4 is calculated using LCOH numbers without any CO2 tax. Therefore, the value of COCA also gives the minimum CO2 emissions tax value needed to make the GSR-H2 process economically more attractive than the conventional SMR plant.

<table>
<thead>
<tr>
<th>Table 3 – Assumptions for fixed and variable operating and maintenance costs and fuel costs.</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Fixed O&amp;M Costs</strong></td>
</tr>
<tr>
<td>Operating Labor</td>
</tr>
<tr>
<td>Maintenance, Support and Administrative Labor</td>
</tr>
<tr>
<td>Property Taxes</td>
</tr>
<tr>
<td>Insurance costs</td>
</tr>
<tr>
<td>NG price (Fuel Cost)</td>
</tr>
<tr>
<td><strong>Variable O&amp;M Costs</strong></td>
</tr>
<tr>
<td>Cooling Water</td>
</tr>
<tr>
<td>Make Up Costs</td>
</tr>
<tr>
<td>Process Water Cost</td>
</tr>
<tr>
<td>Catalysts and Sorbent Replacement</td>
</tr>
<tr>
<td>Oxygen Carrier cost</td>
</tr>
<tr>
<td>WGS catalyst cost</td>
</tr>
<tr>
<td>Adsorbent cost</td>
</tr>
<tr>
<td>$6#(mixture of activated)</td>
</tr>
<tr>
<td>$6#(carbon and Zeolite 5A)</td>
</tr>
<tr>
<td>Replacement Period</td>
</tr>
<tr>
<td>CO2 Transport and $6#Storage Costs</td>
</tr>
<tr>
<td>Emissions Tax (CO2)</td>
</tr>
<tr>
<td>Electricity price</td>
</tr>
<tr>
<td>Steam exports</td>
</tr>
<tr>
<td><strong>Consumables</strong></td>
</tr>
<tr>
<td><strong>$/person-year</strong></td>
</tr>
<tr>
<td><strong>% of TOC</strong></td>
</tr>
<tr>
<td><strong>$/GJ LHV</strong></td>
</tr>
<tr>
<td><strong>$/m 3</strong></td>
</tr>
<tr>
<td><strong>$/kg</strong></td>
</tr>
<tr>
<td><strong>$/m 3</strong></td>
</tr>
<tr>
<td><strong>$/kg [39,40]</strong></td>
</tr>
<tr>
<td><strong>$/ton CO2</strong></td>
</tr>
<tr>
<td><strong>$/ton CO2</strong></td>
</tr>
<tr>
<td><strong>Years</strong></td>
</tr>
<tr>
<td><strong>$/ton</strong></td>
</tr>
<tr>
<td><strong>$/MWh</strong></td>
</tr>
<tr>
<td><strong>$/ton [41]</strong></td>
</tr>
</tbody>
</table>

\[
\text{COCA} \left( \frac{\$}{\text{ton CO}_2} \right) = \frac{\text{LCOH}_{\text{GSR-H2}} - \text{LCOH}_{\text{ref}}}{\text{CO2}_{\text{ref}} - \text{CO2}_{\text{GSR-H2}}}
\]

**Results and discussions**

This section is presented in three parts. The first two parts describe the technical and economic performance of the different GSR-H2 process configurations compared to the SMR benchmark. The third part consolidates the discussion in the first two sub-sections and presents scenarios, challenges and opportunities for deployment of GSR-H2 process in the form of different sensitivity studies. The main results for the techno-economic analysis for the SMR and GSR-H2 processes are presented in Table 4.

**Technical analysis**

The detailed technical performance comparison between the conventional SMR plant and the base case GSR-H2 process with and without added thermal mass was reported in Nazir et al. [17]. Results in this section are more focussed towards improvements in the GSR-H2 process. The conditions in the GSR-H2 process for different cases are shown in Table 5.

**Effect of having a two-phase evaporator (GSR-H2 in case 2)**

Case 2 has 0.93%-point higher equivalent hydrogen production efficiency than the base case GSR-H2 process. In case 2, low temperature heat is utilised in preparing steam in the two-phase evaporator, resulting in lower heat rejection to the cooling water as seen in Table 5. This slightly improved heat recovery allowed for 25 °C greater pre-heating of the fuel and steam to reforming, requiring a little less PSA off-gas combustion to heat the inlet gases in the GSR. This is reflected in a lower air (oxygen) requirement and a 0.35 %-point higher hydrogen production efficiency. Table 5 also shows a higher S/C ratio for this case. This is the result of a lower PSA off-gas requirement that demands more H2 extraction from the syngas which, in turn, requires more steam to increase CH4 conversion in the GSR reactors and CO conversion in the WGS reactors. The reforming step is also slightly longer, leading to a lower average reforming temperature, which requires more steam to increase CH4 conversion. The net electricity consumption in case 2 is 6.6% lower than in base case, mainly due to lower compression work for compressing air (lower air flow to GSR), hydrogen (less pressure drop in the syngas stream due to the absence of NG pre-heater), PSA off-gas (lower PSA off-gas demand) and CO2 stream (using two-phase evaporator cools down the CO2 stream in one step, avoiding higher pressure drops due to multiple coolers as in the base case and case 1). Since a fraction of steam is prepared in the two-phase evaporator in case 2, the heat in N2 stream can be used in the N2-turbine to produce more power when compared to the base case. Therefore, integrating the two-phase evaporator in the GSR-H2 process to raise steam for reforming reduces the efficiency penalty in GSR-H2 process...
from 3.8%-points to 2.9%-points with reference to the conventional SMR plant, reducing the SPECCA from 1.06 to 0.79 MJ/kg-CO2.

Effect of additional thermal mass and two-phase evaporator (GSR-H2 in case 3)
Having additional thermal mass in the GSR results in overall higher temperatures in the reactor. This allows the required fuel conversion to be achieved with a lower S/C ratio, leading to considerably less rejection of the condensation enthalpy of excess steam in the syngas to the cooling water as seen in Table 5. The main benefit of the lower steam requirement is that the N2 stream can be expanded from a high TIT, leading to 74% greater power production from the N2-turbine compared to case 2. Additional heat after the N2-turbine is exported as 6 bar steam. This reduction in electricity consumption and increase in steam exports increase the equivalent hydrogen production efficiency of case 3 by 3.99%-points relative to case 2, even though hydrogen production efficiency is only 0.4%-points higher. This makes case 3 more efficient than the SMR benchmark, resulting in a negative SPECCA value of /C0 0.26 MJ/kg-CO2.

Effect of heat integration with pre-heated air for oxidation step of the GSR (case 4)
Pre-heating air for the oxidation step of the GSR with the N2 stream reduces the amount of PSA off-gas combustion required to heat the air stream to the reactor temperature. This allows more of the heating value in the fuel to be converted to H2 instead of heat and is reflected in a 3.95%-point increase in hydrogen production efficiency compared to case 3. However, the lower PSA off-gas requirement needs greater H2 extraction in the PSA unit, which requires more steam for converting CH4 and CO to H2, hence the increase in S/C ratio relative to case 3. The main trade-off for the increased
hydrogen production efficiency is the low N₂-turbine inlet temperature (TIT) that strongly reduces power production relative to case 3. Hence, case 4 increases H₂ production at the cost of greater net electricity consumption. Relative to case 3, this configuration loses 0.87 %-points of equivalent H₂ production efficiency, but this is only due to the absence of 6-bar steam exports with limited economic value.

**Economic analysis**

Table 4 and Fig. 6 present the main results from the economic analysis of the SMR and GSR-H₂ processes. Fuel cost accounts for the maximum share in the LCOH in both the processes, with 55% of the LCOH in SMR plant and 49–52% of the LCOH in GSR-H₂ cases. The fuel cost element in the LCOH is inversely proportional to the hydrogen production efficiency. In the SMR plant, the capital requirement contributes about 21% of the LCOH and is in a similar range (20–21%) for GSR-H₂ cases that have N₂ turbines with lower TITs (base case, case 2 and 4). In case 1 and 3, higher N₂ turbine power output results in higher turbo-machinery costs as shown in Fig. 7a. The costs associated with heat exchangers is higher in SMR plant because of the lower heat transfer coefficient for gas-gas heat transfer in the pre-heaters that recover heat from the low-pressure exhaust gas from the FTR burners. Case 4 of GSR-H₂ has an additional air pre-heater before the oxidation step of the GSR resulting in higher heat exchanger related capital costs when compared to other GSR-H₂ cases. However, the costs associated with high temperature turbomachinery is relatively high and, therefore, the capital cost element in LCOH for case 1 and 3 is 1–2 %-points higher than the other GSR cases. Fig. 7a also shows that the GSR reactors have a slightly lower cost than the FTR unit in the SMR benchmark case, even after adding a 30% process contingency to GSR. This is due to the simplicity of the GSR reactors relative to the FTR that houses a large number of pressurized reforming tubes in a high temperature furnace. The FOM cost is higher in case 1 and 3 because these costs are calculated as a fixed fraction of capital costs.

The VOM costs in the SMR plant account for 12.4% of the LCOH, whereas it ranges between 13 and 18% for the GSR-H₂ process. The VOM costs in the SMR plant are mainly associated with the emission taxes, whereas electricity and CO₂ transport and storage costs account for the major fraction of the VOM costs in GSR-H₂ process (Fig. 7b). The CO₂ transport and storage costs are similar in all the GSR-H₂ cases since the CO₂ capture rate is similar. However, the VOM costs for cases with higher electrical demand are higher.

For the assumptions described in section Economic analysis methodology, the base case GSR-H₂ plant with ~96% CO₂ capture has a slightly lower LCOH when compared to SMR plant without CO₂ capture. The VOM costs in the base case GSR-H₂ process are higher due to higher net electricity demand, which is compensated by higher hydrogen production efficiency and hence lower fuel costs than the SMR plant. The improvements in cases 1–4 help in reducing the hydrogen production costs from the GSR-H₂ process. Case 4 has the lowest LCOH which is ~4.5% less than the LCOH from SMR plant. Case 3 has the highest equivalent hydrogen production efficiency but has 1.6% higher LCOH than the GSR-H₂ process in case 4, because case 4 produces more hydrogen at the cost of higher overall electricity input. Although the CO₂ emission tax of 23 $/t-CO₂ was assumed to calculate the LCOH, it is shown in Table 4 that an emission tax value of 15 $/t-CO₂ will make the GSR-H₂ process configured according to case 4 economically more attractive than the SMR plant.

Most components of the GSR-H₂ process are technologically mature, implying that uncertainties involved in capital cost estimation using the established commercial tools employed in this study are relatively small. In addition, the GSR-H₂ plants use a process layout with similar process components to the benchmark SMR plant, so any uncertainties in the costs of components like the pre-reformer, water-gas shift reactors and PSA unit will have a limited effect on conclusions from this comparative study. The only process component at a lower technology readiness level is the GSR reactor. In this case, uncertainty is limited by the simplicity of the standalone bubbling fluidized beds employed, so the capital cost estimate with the 30% added process contingency to GSR. This is due to the simplicity of the GSR reactors relative to the FTR that houses a large number of pressurized reforming tubes in a high temperature furnace. The FOM cost is higher in case 1 and 3 because these costs are calculated as a fixed fraction of capital costs.

Fig. 6 – Distribution of different components of levelised cost of hydrogen (LCOH) for SMR and GSR-H₂ process.

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associated with steam exports have minimal impact on the LCOH of the processes.

Finally, an important technical uncertainty that should be mentioned is the performance and durability of high temperature valves downstream of the GSR reactor. If technical constraints from these valves restrict the maximum reactor temperature below the value of 1100 °C assumed in this study, the efficiency of the process will decline as more steam will be needed to achieve sufficient fuel conversion, increasing fuel consumption and cost. On the other hand, valves that can operate at even higher temperature could further improve GSR efficiency. This is an important element that must be demonstrated during scale-up of the GSR technology.

Scenarios for low-cost hydrogen production with CO2 capture using GSR

As discussed in the previous section, fuel cost is the major component of the LCOH in both the SMR and GSR-H2 plants. GSR-H2 processes shift some of this energy input to electricity, making the electricity price another important parameter. In addition, CO2 transport and storage costs are also uncertain with CO2 utilization potentially resulting in low or negative costs, whereas public resistance and insufficient scale can strongly increase costs. Fig. 9 therefore shows the sensitivity of the LCOH and COCA of SMR and GSR-H2 plants to NG price, electricity price and CO2 transport and storage costs. The COCA presented in Fig. 9 is equivalent to the minimum CO2 emission tax needed to make the GSR-H2 process more economically attractive than the SMR plant.

The SMR and GSR-H2 processes are highly sensitive to the NG price as seen in Fig. 9a. From the reference price of NG (6.9 $/GJ-LHV), a change in NG price by 2 $/GJ-LHV affects the LCOH in SMR plant by 16% and in the GSR-H2 plant by 14–15%. Since the GSR plants displace some NG energy input with electricity, they are less sensitive to increases in NG price. This also explains the reduction in COCA as NG price is increased (Fig. 9b). At a very high NG price of 9 $/GJ-LHV, the GSR-H2 (case 4) process will need only a CO2 emission tax of 8 $/t-CO2 for the GSR-H2 process with >96% CO2 capture to perform economically better than the SMR plant without capture.

Fig. 7 – Split of the a) Bare erected cost b) Variable O&M costs.

Fig. 8 – Effect of oxygen carrier lifetime on levelised cost of hydrogen from GSR-H2 process.

Fig. 9c shows the sensitivity of the LCOH of SMR and GSR-H2 plants with change in electricity price. The low power consumption of the SMR plant makes it insensitive to the electricity price. However, the LCOH of the GSR-H2 plant varies between 2 and 4% when electricity price changes by 20 $/MWh around the base value of 69 $/MWh. This difference in sensitivity is best exemplified by comparing cases 3 and 4. The trade of more H2 production for greater electricity consumption in case 4 makes it more sensitive to electricity price variations. As expected, Fig. 9d shows that higher electricity prices substantially increase the COCA for all GSR plants. Although not directly studied here, it can be noted that COCA will also vary with the source of electricity, which influences the indirect CO2 emissions of the GSR plant. The effect of electricity source on the CO2 avoidance rate was discussed in Nazir et al. [17]. In general, GSR will be most attractive when clean electricity can be bought from the grid at a low cost. This should be an important consideration when considering investment in GSR-H2 plants.
The CO₂ transport and storage costs depend on the type of transport and also the scale of flow being transported [42]. Fig. 9e shows the sensitivity of the LCOH of GSR-H₂ plants with the CO₂ transport and storage costs. If the transport and storage costs are doubled, the LCOH of the GSR-H₂ processes increases by 4%. Naturally, COCA increases in direct proportion with the CO₂ transport and storage cost (Fig. 9f).

When considering the range of COCA values in Fig. 9, it is clear that all three of these variables have a significant impact on the competitiveness of GSR-H₂ against conventional SMR. However, over the ranges investigated, the electricity price
has the largest impact, with the natural gas price and CO₂ transport and storage costs showing a slightly lower sensitivity.

Fig. 9 showed the sensitivity of the LCOH and COCA of SMR and GSR-H₂ plants with the important future energy market drivers like NG price, electricity price and CO₂ transport and storage costs. However, the future energy scenario is complex and will depend on multiple factor interaction. Table 6 presents twelve different future energy scenarios that reflect the change in NG price, electricity price and stricter emission regulations through a CO₂ tax. The NG price is location dependent, for example ~3 $/GJ-LHV in USA and 6–8 $/GJ-LHV in Europe [43] and is projected to increase in the future energy scenarios [28,44]. The electricity price from thermal plants is sensitive to the fuel price, and with CCS increases the price by 15–30% [44]. However, the electricity generation sector is undergoing a major shift towards renewables. This shift has brought down the cost of electricity from renewables drastically in the last 10 years [45]. Clean energy transitions also depend on policy measures to control emissions. For example, the CO₂ emission tax is projected to increase with stricter guidelines for industries to reduce their emissions. Scenarios 1–3 and 7–9 in Table 6 depict scenarios with low NG price (for example: USA and the Middle East), with scenarios 1–3 having low electricity price (for example: countries with abundant local fossil fuel reserves like the Middle East and/or renewable energy potential like Norway) and scenarios 7–9 having high electricity prices (for example: fossil fuel importers with limited renewable energy potential like Japan). The scenarios also show the effect of increasing CO₂ emission taxes. Scenarios 4–6 and 10–12 depict scenarios with high NG price (for example: NG importers like Europe, Japan and most of developing Asia) factoring in low and high electricity price and CO₂ emission tax.

The LCOH from the SMR benchmark and the two most promising GSR-H₂ configurations (cases 3 and 4) is calculated for the twelve scenarios (Table 6) and shown in Fig. 10. Fig. 10 also shows the LCOH for an additional case: case 4 without CO₂ capture. As outlined in Nazir et al. [17], this is a promising commercialization pathway for the GSR technology. Constructing the plant without CO₂ capture can outcompete current SMR technology in an environment without significant CO₂ taxes and such a plant will be very simple to retrofit for CO₂ capture at a later stage when CO₂ taxes are higher and CO₂ transport and storage infrastructure is in place.

Fig. 10 clearly shows that case 4, either with or without CO₂ capture, is the lowest cost option in all but three of the 12 scenarios. This illustrates the robustness of the GSR-H₂ business case when the retrofitting pathway described above is followed. It is only in the scenarios combining low natural gas prices with high electricity prices where the relatively high electricity consumption of case 4 reduces its competitiveness. In these scenarios, the SMR plant is the cheapest at low CO₂ prices and case 3 is cheapest at high CO₂ prices. It can also be noted that such a combination of low natural gas prices and high electricity prices is unlikely to persist for extended periods because cheap natural gas will lead to a rapid expansion of low-cost electricity from natural gas combined cycle power plants (e.g. the shale revolution in the US).

Finally, it can be mentioned that successful commercialization of GSR-H₂ plants on the scale investigated in this study (10 TPH of NG) can lead to widespread deployment of GSR combined cycle power plants for flexible power and hydrogen production at a scale of 100–150 TPH of NG [27]. Such flexible GSR plants can significantly reduce overall power system costs and emissions in a future scenario with high shares of wind and solar power [46].

### Conclusions

This study presented a techno-economic assessment of the GSR-H₂ plant for hydrogen production from natural gas reforming with inherent CO₂ capture. Improved process configurations were devised to eliminate the energy penalty of CO₂ capture and an economic assessment was completed to assess whether the high process efficiency translates into competitive hydrogen production costs.

The GSR-H₂ process achieved CO₂ avoidance costs as low as 15 $/ton, making it a promising candidate for affordable clean hydrogen production. Such advanced reforming processes are generally not considered in long-term hydrogen production scenarios.
pathways such as in the recent IEA “Future of Hydrogen” [4] report where CO2 capture from steam methane reforming plants remains expensive in longer-term scenarios. It is recommended that such high-level reports acknowledge the possibility of larger long-term cost reductions for natural gas reforming with CO2 capture similar to the current cost reduction assumptions for electrolysis.

The most attractive commercialization pathway for the GSR-H2 technology is to first construct the plant without any CO2 capture. In this configuration, hydrogen production costs are clearly lower than that of the SMR benchmark. When CO2 taxes eventually increase and CO2 transport and storage infrastructure becomes available, the plant can easily and cheaply be retrofitted for CO2 capture to ensure decades of continued profitable operation. This pathway makes the GSR-H2 plant insensitive to uncertainties in the future development of CO2 taxation schemes.

Oxygen carrier lifetime is the key technical uncertainty that can adversely affect GSR performance. Lifetimes below 1 year start to significantly increase the hydrogen production cost. For this reason, long-term demonstration of oxygen carrier stability under realistic GSR operating conditions should be the first priority for scale-up and commercialization of this promising clean energy technology. Following this demonstration, the simplicity of the GSR reactor design can allow for rapid scale-up to commercial scale.

When considering operating costs, a distinguishing feature of GSR-H2 is that it exchanges some fuel consumption for electricity consumption. Relative to conventional SMR, this makes the GSR-H2 plant more suitable to regions with higher fuel costs and/or lower electricity prices. The trade-off between fuel and electricity consumption can be controlled with small changes to the GSR-H2 process layout to adapt the plant to application in different world regions.

Based on all these features of the GSR-H2 process, further scale-up and demonstration activities can be recommended.

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REFERENCES


