

Regional Assessment of Hydrogen Production and Use in the Intermountain West United States

Prashant Sharan ^{*,†}, Lucky E. Yerimah [†], Manvendra Dubey, Harshul Thakkar, Mohamed Mehana, Troy Semelsberger, Michael Heidlage and Rajinder Singh

Los Alamos National Laboratory, Bikini Atoll Road, Los Alamos, NM 87545, USA;
eshemomoh@lanl.gov (L.E.Y.); dubey@lanl.gov (M.D.); harshul@daircapture.com (H.T.);
mzm@lanl.gov (M.M.); troy@lanl.gov (T.S.); mheidlage@lanl.gov (M.H.); rsingh@lanl.gov (R.S.)

* Correspondence: prashant.sharan@lanl.gov

† These authors contributed equally to this work.

S1.

S1.1. Model Description

In this section, we describe a general procedure to calculate the mass flowrate of different gases flowing through the SMR, WGS, PSA reactors and combustion chamber.

Typically, NG is >90% methane (by mass), with heavier hydrocarbons present in low fractions. The typical composition of NG is given in Table S1. The higher hydrocarbons present in NG undergo reforming similar to methane, and the reactions are:

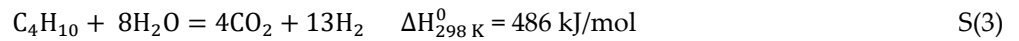
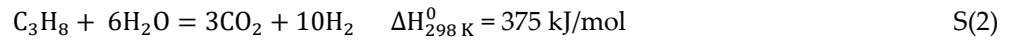
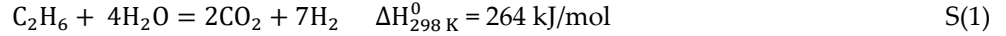


Table S1. Typical pipeline composition of NG flowing through an interstate pipeline [55].

	Mole Fraction	Mass Fraction
Methane	94.90%	90.18%
Ethane	2.50%	4.45%
Propane	0.20%	0.52%
iso-butane	0.03%	0.10%
butane	0.03%	0.10%
Iso-pentane	0.01%	0.04%
pentane	0.01%	0.04%
Hexanes	0.01%	0.04%
Nitrogen	1.60%	2.66%
Carbon dioxide	0.70%	1.83%
Oxygen	0.02%	0.02%
Hydrogen	0.00%	0.00%

Table S2. Mass, energy, and momentum balance equations of the SMR and WGS models.

Mass Balance

$$\frac{\partial C_i}{\partial t} - \frac{\partial}{\partial z} \left(D_L \frac{\partial C_i}{\partial z} \right) + \frac{\partial(u_z C_i)}{\partial z} + (1 - \epsilon_b) \rho_s \sum_{j=1}^{Nr} v_{ij} \eta_j r_j = 0$$

where r_j represents the reaction rates for SMR and WGS, $i = 1 \cdots C - 1$

Energy Balance

$$\rho_g C_{pg} \frac{\partial T}{\partial t} - \frac{\partial}{\partial z} \left(\lambda \frac{\partial C_i}{\partial z} \right) + \rho_g C_{pg} \frac{\partial(u_z T)}{\partial z} + \frac{4}{D} U(T_w - T) + (1 - \epsilon_b) \rho_s \sum_{j=1}^{Nr} \Delta H_r \eta_j r_j = 0$$

Momentum Balance

$$-\frac{\partial P}{\partial z} = \frac{150\mu(1 - \epsilon_b)^2}{\epsilon_b^3 d_p^2} u_z + \frac{1.75(1 - \epsilon_b)\rho_g}{\epsilon_b^3 d_p} u_z^2$$

The conversion efficiency for the SMR process is typically less than 100%. This leaves behind unused NG in the feed stream to flow into the WGS reactor and PSA separator. The generalized form for net reaction taking place in SMR with a natural gas molar flow rate of F_{NG} , with a conversion efficiency of x leaving the reactor, is given as:

$$\begin{aligned} SMR_{out} = & x[F_{NG,CH_4} + 2F_{NG,C_2H_6} + 3F_{NG,C_3H_8} + 4F_{NG,C_4H_{10}}]_{CO} \\ & + x[3F_{NG,CH_4} + 5F_{NG,C_2H_6} + 7F_{NG,C_3H_8} + 9F_{NG,C_4H_{10}}]_{H_2} \\ & + x[(3 - x)F_{NG,CH_4} + (5 - 2x)F_{NG,C_2H_6} + (7 - 3x)F_{NG,C_3H_8} + (9 - 4x)F_{NG,C_4H_{10}}]_{H_2O} \\ & + (1 - x)[F_{NG,CH_4} + F_{NG,C_2H_6} + F_{NG,C_3H_8} + F_{NG,C_4H_{10}}] + [F_{NG,CO_2}]_{CO_2} \end{aligned} \quad S(4)$$

The reformed natural gas undergoes WGS reaction in two stages, at ca. 350 °C and 200 °C. The reformed gas leaving the SMR reactor is at a high temperature (700 °C) and needs to be cooled before flowing to the WGS reactor. For WGS conversion efficiency of y , the composition of the syngas flowing out of the WGS is given as:

$$\begin{aligned} WGS_{out} = & x(1 - y)[F_{NG,CH_4} + 2F_{NG,C_2H_6} + 3F_{NG,C_3H_8} + 4F_{NG,C_4H_{10}}]_{CO} \\ & + x[(3 + y)F_{NG,CH_4} + (5 + 2y)F_{NG,C_2H_6} + (7 + 3y)F_{NG,C_3H_8} + (9 + 4y)F_{NG,C_4H_{10}}]_{H_2} \\ & + x[(3 - x - y)F_{NG,CH_4} + (5 - 2x - 2y)F_{NG,C_2H_6} + (7 - 3x - 3y)F_{NG,C_3H_8} + (9 - 4x - 4y)F_{NG,C_4H_{10}}]_{H_2O} \\ & + (1 - x)[F_{NG,CH_4} + F_{NG,C_2H_6} + F_{NG,C_3H_8} + F_{NG,C_4H_{10}}] \\ & + [xy(F_{NG,CH_4} + 2F_{NG,C_2H_6} + 3F_{NG,C_3H_8} + 4F_{NG,C_4H_{10}}) + F_{NG,CO_2}]_{CO_2} \text{ mol/s} \end{aligned} \quad S(5)$$

The shifted syngas is cooled close to 40 °C before flowing in the PSA reactor for H_2 separation from the syngas. For H_2 recovery of R_{H_2} the net amount of H_2 generated is:

$$H_{2,production} = R_{H_2} x[(3 + y)F_{NG,CH_4} + (5 + 2y)F_{NG,C_2H_6} + (7 + 3y)F_{NG,C_3H_8} + (9 + 4y)F_{NG,C_4H_{10}}]_{H_2} \text{ mol/s} \quad S(6)$$

The trail gas composition leaving the PSA unit flows into the reformer combustion chamber, where it is combusted with additional natural gas. The composition of tail gas leaving the PSA is:

$$\begin{aligned}
PSA_{out} = & x(1-y)[F_{NG,CH_4} + 2F_{NG,C_2H_6} + 3F_{NG,C_3H_8} + 4F_{NG,C_4H_{10}}]_{CO} \\
& + (1-R_{H_2})x[(3+y)F_{NG,CH_4} + (5+2y)F_{NG,C_2H_6} + (7+3y)F_{NG,C_3H_8} + (9+4y)F_{NG,C_4H_{10}}]_{H_2} \\
& + (1-x)[F_{NG,CH_4} + F_{NG,C_2H_6} + F_{NG,C_3H_8} + F_{NG,C_4H_{10}}] \\
& + [yF_{NG,CH_4} + 2yF_{NG,C_2H_6} + 3yF_{NG,C_3H_8} + 4yF_{NG,C_4H_{10}} + F_{NG,CO_2}]_{CO_2} \text{ mol/s}
\end{aligned} \tag{S(7)}$$

SMR is an endothermic reaction, requiring heat in the temperature range of 800–1000 °C, which is supplied by a mixture of recycled tail gas from PSA unit and natural gas. For additional heat supply, the NG can directly be supplied to the reformer combustion chamber and is given as:

$$NG_{combustion} = \frac{\dot{E}_{heat} - PSA_{out,i}\Delta H_i}{\Delta H_{NG}} \tag{S(8)}$$

where $\dot{E}_{heat}$ is the SMR heating energy required, ΔH_i is the heat of combustion for reactants i (H_2 and unused natural gas).

S1.2. Energy Calculation for SMR Process

Figure S1, shows the compositive curve for steam methane reformer. Process 1–4 represents pre-heating for water and natural gas. Process 2–3 represents evaporation of water, where the water gets converted to steam. The steam and natural gas is further pre-heated (process 3–4), before entering into the SMR reactor. The entry temperature to the SMR reactor is fixed at 530 °C. The temperature of the gases gradually increases to desired temperature of 850 °C in the SMR reactor due to the heat supplied from the combustion (process 4–5). Simultaneously the reforming of natural gas occurs in the SMR reactor. The syngas generated at 850 °C is first cooled to 350 °C, (process 6–7) before flowing into the WGS reactor. The WGS reactor is an exothermic process, and is assumed to be operating at constant temperature (process 7–8). The syngas coming out from the WGS is then cooled to 200 °C (process 8–9), before flowing into the second WGS, where any leftover CO gets converted to CO_2 (process 9–10). The syngas is then cooled to 40 °C before flowing into the PSA process for H_2 purification (process 10–13). Process 11–12 represents the steam condensation process.

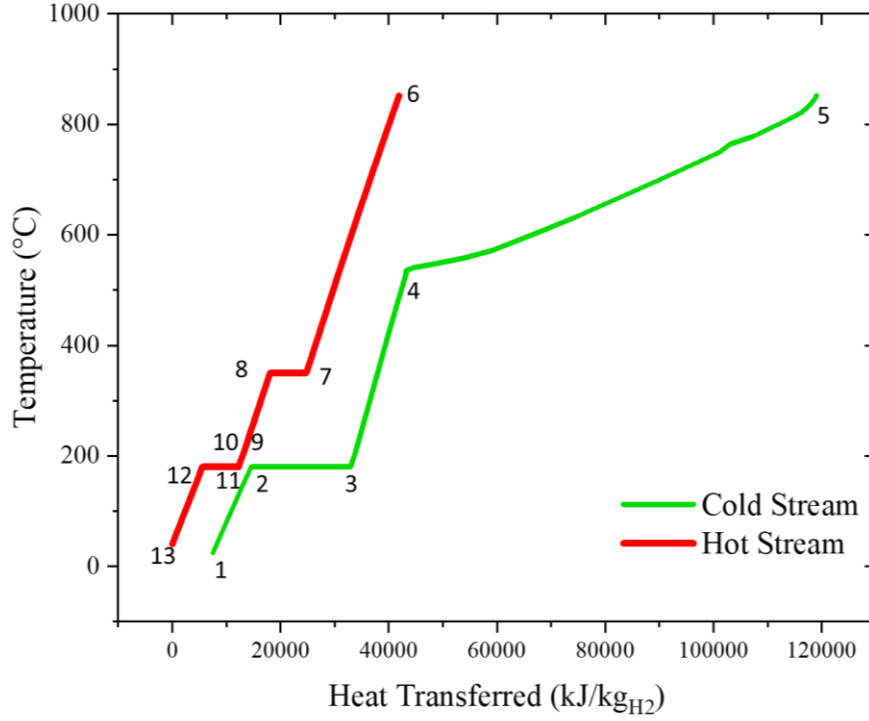


Figure S1. Composite curve for steam methane reformer process.

The distance between 6–7, shown by Q_{hot} represents the net heating required by the SMR process, similarly the 11-1 shows the cooling required $Q_{cold, SMR}$. The net energy required for H_2 production $\dot{E}_{heat}$ is given as:

$$\dot{E}_{heat} = \frac{Q_{hot}}{\dot{m}_{H_2} \times \eta_{SMR}} \quad S(9)$$

Using Equation S(5), the additional amount of natural gas for combustion can be calculated. The flue gas coming out from the SMR combustion chamber is close to 1000 °C. The flue gas energy can be utilized for preheating the air, natural gas and tail gas. By recovering the flue gas energy for pre-heating, the flue gas can be cooled to 420 °C. The flue gas needs to be cooled before emitting it to the environment ($Q_{cool, flue gas}$). The net cooling required for the process is given as:

$$\dot{E}_{cool} = \frac{Q_{cold, SMR} + Q_{cool, flue gas}}{\dot{m}_{H_2}} \quad S(10)$$

S1.3. Global Warming Potential for Natural Gas

Table S1, list the global warming potential (GWP) for different constituents of natural gas for 100-years. Methane being the major constituents at 90.2%, and with a maximum GWP of 28 $kg_{CO_2,eq}/kg_{CH_4}$, the net GWP for natural gas comes out to be 25.8 $kg_{CO_2,eq}/kg_{NG}$. It may be noted the GWP for heavier hydrocarbon from butane, pentane and hexane is assumed to be the same at 7 $kg_{CO_2,eq}/kg_{gas}$ [105]. The other assumption made is that all the gases leaks are in same proportion.

Table S3. Global warming potential for different constituent of natural gas.

Constituents	Mass Fraction [55]	GWP (100-years) [105,106]
Methane	90.18%	28
Ethane	4.45%	10.2
Propane	0.52%	9.8
iso-butane	0.10%	7
butane	0.10%	7
Iso-pentane	0.04%	7
pentane	0.04%	7
Hexanes	0.04%	7
Nitrogen	2.66%	0
Carbon dioxide	1.83%	1
Oxygen	0.02%	0
Hydrogen	0.00%	0
Natural Gas	100%	25.8

S2. Energy Consumption

S2.1. Compression Energy Required

The NG or CO₂ enters the compressor at temperature T_{in} and pressure P_{in} , with net enthalpy h_{in} . The compressor increases the pressure of the working fluid from P_{in} to P_{out} . If compression process is isentropic, i.e., the entropy of inlet stream will be equal to the outlet stream. In such a case, the enthalpy is given as $h_{out,s}$. However, for actual compressor process is non-isentropic, the net work for natural gas and CO₂ is given as [77]:

$$\dot{w}_{comp} = \frac{(h_{out,s} - h_{in})}{\eta_{isen,comp}} \quad S(11)$$

where $\eta_{isen,comp}$ is the compressor isentropic efficiency and is assumed to be 80% [107]. The outlet enthalpy for the gas leaving the compressor is given as:

$$h_{out} = h_{in} + \dot{w}_{comp} \quad S(12)$$

Using the outlet enthalpy and pressure, the temperature of the gas leaving the compressor can be calculated. Often an upper limit on operating temperature is recommend for the safe working of the compressor. If the temperature exceeds the recommended limit the pressure ratio needs to be reduced. For present case we have limited the temperature rise of the gases 150 °C. To achieve higher compression, the gas needs to be cooled to lower temperature via air cooler before sending it to the next stage for compression. We have assumed the inlet temperature of natural gas entering the compressor is at 40 °C, while the cooling air temperature is 25 °C.

Table S4. Compressor energy required for natural gas.

Stages	Pressure Ratio	Operating Pressure (bar)	Work Done (kWh _e /kg _{NG})	Cooling (kWh _{th} /kg _{NG})
1	3.17	9.50	0.070	0.072
2	2.80	26.60	0.061	0.066

3	2.72	72.41	0.058	0.072
Total	24.14	72.41	0.19	0.21

Typical natural gas coming out from oil and gas field is at 3 bar pressure. Table S2 shows the net electrical and cooling energy required by different stages. The net work done for compression is 0.19 kWh_e/kg_{NG}, while the cooling required is 0.21 kWh_{th}/kg_{NG}. It is very common to use air cooler for cooling the compressor. Using a pressure drop of 200 Pa for air compressor, the net electrical energy required for air cooler is 0.003 kWh_e/kg_{NG}.

For compressing H₂ to 180 bars, requires 1.63 kWh_e/kg_{H2} for compression and 0.02 kWh_e/kg_{H2} for cooling. For SMR_{CCS} the CO₂ generated needs be transported and sequestered at supercritical conditions (>71 bar pressure and >31 °C). This needs CO₂ compression to 100 bar pressure, which requires 0.11 kWh_e/kg_{CO2} for compression and 0.002 kWh_e/kg_{CO2}.

S2.2. Energy Required for H₂ Production

Table S3 shows the process parameter for H₂ production. Using the process parameter, the composite curve can be generated and is shown in Figure 3. Table S4 shows the energy required for H₂ production. The SMR requires around 23.8 kWh_{th}/kg_{H2} of heating energy; however, due to H₂ leak in compressor the net SMR energy required increases to 24.1 kWh_{th}/kg_{H2}, while for higher H₂ leak for regional H₂ the net SMR energy required increases to 25.4 kWh_{th}/kg_{H2}. Electrical energy is required for compression of natural gas to pump it through the pipelines, H₂ compression to 180 bar pressure, water treatment and pumping, and operating the cooling tower. For CCS thermal energy is required for regeneration of MEA, electrical energy for pumping and CO₂ compression for sequestration. The thermal energy required for SMR is 24.1 kWh_{th}/kg_{H2} while the electrical energy required is 2.5 kWh_e/kg_{H2}. The energy required increases for SMR with carbon capture to 35.1 kWh_{th}/kg_{H2} and 4.3 kWh_e/kg_{H2} and for regional H₂ production to 36.3 kWh_{th}/kg_{H2} and 3.9 kWh_e/kg_{H2}.

Table S5. Stream flow data for the SMR process.

	Stream	Temperature		Flowrate (kg/s)					
		T _{in} (°C)	T _{out} (°C)	H ₂ O	NG	H ₂	CO	CO ₂	Pure H ₂
Feed pre-heating	1–2	25.0	179.9	9.1	2.8				
Feed pre-heating	2–3	179.9	179.9	9.1	2.8				
Feed pre-heating	3–4	179.9	531.2	9.1	2.8				
SMR and feed heating	4–5	531.2	535.6	9.1	2.7	0.0	0.1		
SMR and feed heating	4–5	535.6	540.4	9.0	2.7	0.0	0.2		
SMR and feed heating	4–5	540.4	549.5	8.8	2.5	0.1	0.5		
SMR and feed heating	4–5	549.5	558.4	8.7	2.4	0.1	0.7		
SMR and feed heating	4–5	558.4	571.6	8.6	2.3	0.2	0.9		
SMR and feed heating	4–5	571.6	584.5	8.4	2.1	0.2	1.1		
SMR and feed heating	4–5	584.5	609.3	8.2	1.9	0.3	1.5		
SMR and feed heating	4–5	609.3	632.9	8.0	1.7	0.4	1.8		
SMR and feed heating	4–5	632.9	655.3	7.8	1.5	0.4	2.1		
SMR and feed heating	4–5	655.3	676.5	7.6	1.4	0.5	2.4		
SMR and feed heating	4–5	676.5	696.5	7.4	1.2	0.6	2.7		

SMR and feed heating	4–5	696.5	715.2	7.2	1.1	0.6	3.0	
SMR and feed heating	4–5	715.2	732.8	7.1	0.9	0.7	3.2	
SMR and feed heating	4–5	732.8	749.1	7.0	0.8	0.7	3.3	
SMR and feed heating	4–5	749.1	764.3	6.8	0.7	0.8	3.6	
SMR and feed heating	4–5	764.3	778.2	6.7	0.5	0.8	3.8	
SMR and feed heating	4–5	778.2	790.8	6.6	0.4	0.8	4.0	
SMR and feed heating	4–5	790.8	802.3	6.5	0.4	0.9	4.1	
SMR and feed heating	4–5	802.3	812.6	6.4	0.3	0.9	4.3	
SMR and feed heating	4–5	812.6	821.6	6.3	0.2	0.9	4.3	
SMR and feed heating	4–5	821.6	829.5	6.3	0.2	0.9	4.4	
SMR and feed heating	4–5	829.5	836.1	6.3	0.2	0.9	4.4	
SMR and feed heating	4–5	836.1	841.5	6.3	0.2	0.9	4.5	
SMR and feed heating	4–5	841.5	845.7	6.2	0.2	1.0	4.5	
SMR and feed heating	4–5	845.7	848.7	6.2	0.1	1.0	4.5	
SMR and feed heating	4–5	848.7	850.5	6.2	0.1	1.0	4.5	
SMR and feed heating	4–5	850.5	852.0	6.2	0.1	1.0	4.5	
Syngas cooling	6–7	852.0	350.0	6.2	0.1	1.0	4.5	
WGS 1	7–8	350.0	350.0	3.4	0.1	1.3	0.0	7.1
Syngas cooling	8–9	350.0	200.0	3.4	0.1	1.3	0.0	7.1
WGS 2	9–10	200.0	200.0	3.4	0.1	1.3	0.0	7.1
Syngas cooling	10–11	200.0	179.9	3.4	0.1	1.3	0.0	7.1
Condensation	11–12	179.9	179.9		0.1	1.3	0.0	7.1
Cooling	12–13	179.9	40.0		0.1	1.3	0.0	7.1
PSA	13–14	40.0	40.0		0.1	0.3	0.0	7.1
								1.0

Table S6. Net energy consumption for H₂ production.

	SMR	SMR+CCS	SMR+CCS+RE	Regional SMR+CCS
SMR heating energy (kWh _{th} /kg _{H2})	24.14	24.14	24.14	25.35
H ₂ compression (kWh _e /kg _{H2})	1.65	1.65	1.65	1.73
NG compression (kWh _e /kg _{H2})	0.76	0.93	0.58	0.37
Water Treatment and pumping (kWh _e /kg _{H2})	0.01	0.01	0.01	0.01
Cooling (kWh _e /kg _{H2})	0.11	0.29	0.22	0.29
Carbon capture thermal energy (kWh _{th} /kg _{H2})		10.95	6.49	10.95
Carbon capture electrical energy (kWh _e /kg _{H2})		0.21	0.12	0.21
CO ₂ compression for sequestration (kWh _e /kg _{H2})		1.25	0.77	1.25
Total thermal energy (kWh _{th} /kg _{H2})	24.14	35.09	30.64	36.30

Total electrical energy (kWh _e /kg _{H2})	2.54	4.33	3.35	3.86
--	------	------	------	------

With optimal energy integration and waste heat recovery, the SMR heating energy consumption can be significantly reduced. With novel SMR design, the SMR can be operated at lower temperature, 700 °C, with conversion efficiency close to 100% [41]. Similarly, the hydrogen can be purified with novel membrane process and electrochemical cell having recovery ratio >95% [108]. For the optimized process, the net energy consumption for SMR is 10.3 kWh_e, and the composite curve is shown in Figure S2. The energy breakdown for optimized scenario is shown in Table S5. The electrical energy required reduced to 2.3 kWh_e/kg_{H2}. With carbon capture, the SMR net thermal energy required is 19.4 kWh_{th}/kg_{H2} while the electricity requirement is 3.8 kWh_e/kg_{H2}. For regional H₂ energy required is 19.9 kWh_{th}/kg_{H2} and electricity requirement of 3.4 kWh_e/kg_{H2}.

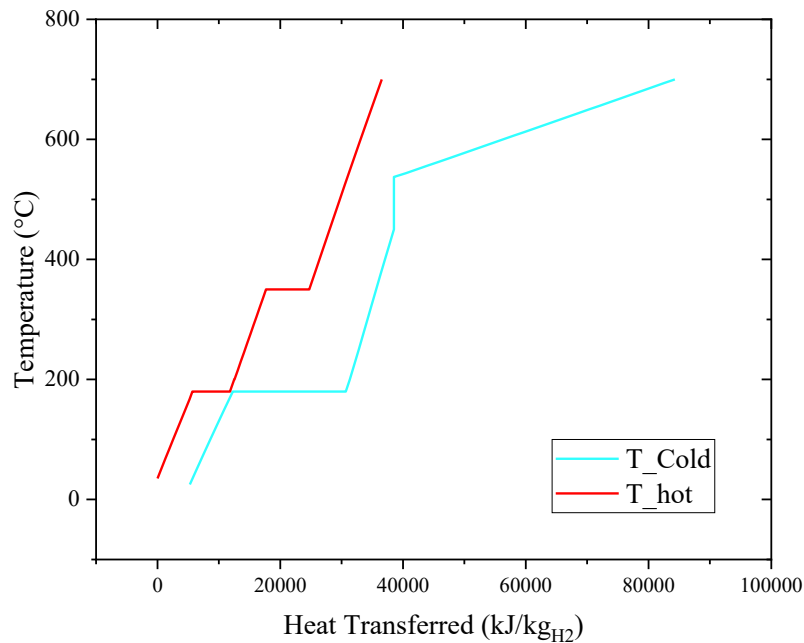


Figure S2. Composite curve for steam methane reformer process with pinch temperature of 10 °C [41].

Table S7. Net energy consumption for H₂ production with optimized scenario.

	SMR	SMR+CCS	SMR+CCS+RE	Regional SMR+CCS
SMR heating energy (kWh _{th} /kg _{H2})	10.36	10.36	10.36	10.87
H ₂ compression (kWh _e /kg _{H2})	1.65	1.65	1.65	1.73
NG compression (kWh _e /kg _{H2})	0.58	0.71	0.45	0.28
Water treatment and pumping (kWh _e /kg _{H2})	0.01	0.01	0.01	0.01
Cooling (kWh _e /kg _{H2})	0.07	0.22	0.16	0.22
Carbon capture thermal energy (kWh _{th} /kg _{H2})		9.02	5.44	9.02

Carbon capture electrical energy (kWh _e /kg _{H2})		0.17	0.10	0.17
CO ₂ compression for sequestration (kWh _e /kg _{H2})		1.03	0.65	0.96
Total thermal energy (kWh _{th} /kg _{H2})	10.36	19.38	15.79	19.90
Total electrical energy (kWh _e /kg _{H2})	2.30	3.79	3.02	3.37

S3. Life Cycle Analysis for H₂ Production

Using Equation S(7)–S(11), for a natural gas flowrate of 1 mol/sec and SMR conversion efficiency of 95%, WGA conversion efficiency of 99.6% and PSA H₂ recovery of 78%, 2.96 mol/sec of pure H₂ is generated. For SMR-WGS-PSA process alone is 7.1 kg_{CO2}/kg_{H2} is generated while 2.8 kg_{NG}/kg_{H2} is required. The tail gas leaving PSA flows at 0.83 mol/sec of H₂, 0.046 mol/sec of CH₄ and 0.001 mol/sec of C₂H₆ for 1 mol/sec of NG flowing in the SMR reactor. For 24.1 kWh_{th}/kg_{H2} heat energy required by the SMR process, 47% is supplied from tail gas burning and additional 0.98 kg_{NG}/kg_{H2} is required to supply heat to SMR process. Due to NG leak, the net amount of NG required increase to 4 kg_{NG}/kg_{H2}. Tail gas combustion generates 0.4 kg_{CO2}/kg_{H2} while natural gas consumption generates 2.8 kg_{CO2}/kg_{H2}. The CO₂ generated due to SMR process and the combustion is 10.4 kg_{CO2}/kg_{H2}. The purified H₂ generated is compressed to 180 bar, which requires 1.65 kWh_e/kg_{H2}, generating 0.9 kg_{CO2}/kg_{H2}, while deionized water generates 0.008 kg_{CO2}/kg_{H2}. The net CO₂ emission SMR process is 11.4 kg_{CO2}/kg_{H2}. 1% H₂ leak from compressor contributes to 0.08 kg_{CO2}/kg_{H2} and due to additional H₂ generation the CO₂ emission increases to 11.65 kg_{CO2}/kg_{H2}. Due to NG leak of 3.5%, the net NG required increases to 4 kg_{NG}/kg_{H2}, of which 0.14 kg_{NG}/kg_{H2} leaks to the environment contributing to 3.5 kg_{CO2}/kg_{H2}. The natural gas extraction generates 1.3 kg_{CO2}/kg_{H2}, while compression NG 80 bar pressure generates additional 0.4 kg_{CO2}/kg_{H2}. The net emission from H₂ generated from SMR process is 16.9 kg_{CO2}/kg_{H2} and is shown in Table S6. 42% of the emissions is from combined SMR+WGS+PSA process, 21% due to NG fugitive emissions, 18% from NG combustion, 8% from NG extraction, 6% from H₂ compression, 3% from tail gas combustion, 2% from NG compression, <1% from H₂ leak and water treatment.

For H_{2,SMR} SMR+combustion process contributes to 10.4 kg_{CO2}/kg_{H2}. With 90% carbon capture, 9.3 kg_{CO2}/kg_{H2} can be captured. The carbon capture would require additional 0.66 kg_{NG}/kg_{H2} to supply heat for CO₂ capture from SMR+combustion. The burning of NG will generate additional CO₂. Here we are trying to capture the CO₂ generated from direct NG burning as well. To capture additional CO₂ requires additional 0.18 kg_{NG}/kg_{H2}. Due to CO₂ captured from SMR and NG flue gas, the net CO₂ captured increases to 11.6 kg_{CO2}/kg_{H2}. While the CO₂ not captured from SMR and NG flue gas is 1.3 kg_{CO2}/kg_{H2}. The net natural gas needed is around 3.8 kg_{NG}/kg_{H2}. The other CO₂ contributing factors are listed in Table S6, with net CO₂ emission from H_{2,SMR+CCS} to 9.5 kg_{CO2}/kg_{H2}.

Table S8. Life cycle analysis for H₂ production for 3.5% NG leak, 1% H₂ leak from compressor while the SMR energy heating energy requirement of 24.1 kWh_{th}/kg_{H2}.

	H _{2,SMR}	H _{2,SMR+CC}	H _{2,SMR+CC+RE}	Direct Methane Burning
NG extraction/processing	1.30	1.58	0.98	0.96
NG compression	0.41	0.50	0.31	0.29
NG leak	3.45	4.21	2.61	2.65

SMR + Combustion	10.39			8.13
H ₂ compression	0.88	0.88	0.03	
H ₂ leak	0.08	0.08	0.08	
CO ₂ not captured		1.28	0.76	
Electrical Energy	0.01	0.14	0.01	
CO ₂ compression		0.68	0.01	
RE for SMR			0.13	
RE for CC			0.23	
CO ₂ released/kg _{H2}	16.51	9.35	5.17	12.03

All units are in kgCO₂/kg_{H2}.

S4. Sensitivity Analysis

S4.1 Gas Leakage

Natural gas leakage is a function of extraction, compression, transportation, and storage [61,70,71]. Different values for NG leakage have been reported in literature. Here we have varied the NG leakage from 0–7%. For 100-year lifetime the H_{2,SMR+CCS} gives superior performance to direct NG burning and is 9% lower than direct burning of NG even at 7% NG leakage. Reducing the NG leakage from 3.5% to 1%, reduces the net CO₂ emissions for H_{2,SMR+CCS} by 32% [see Figure S3 (a)]. Therefore, the focus should be to minimize the NG leakage, as the penalty due to NG leak can be severe.

S4.2. SMR energy consumption

SMR is an energy intensive process with energy consumption of 24.1 kWh_{th}/kg_{H2}. For SMR stoichiometric ratio of steam and methane required is two, however, to improve the reaction kinetics and conversion ratio often steam to methane ratio is varied between three-four. This increases the heat requirement for SMR process, due to increase in the energy required for water evaporation. Therefore, to improve the energy efficiency of the system heat integration becomes essential, particularly for feed preheating. The heat released from water gas shift reaction (exothermic reaction) and cooling of gas streams before it enters the PSA can also be used as a heat source for feed preheating.

With optimal system design and process integration the SMR energy requirement can be brought down to 10.3 kWh_{th}/ kg_{H2}. The heat for SMR process is supplied from the tail gas and natural gas burning. However, due to lower H₂ recovery of 80% from PSA, the net energy required can be reduced to only 11 kWh_{th}/kg_{H2}. Reducing the SMR energy consumption from 30 to 10 kWh_{th}/kg_{H2} can reduce the net CO₂ emission by 40% for H_{2,SMR+CCS}. While with H_{2,SMR+CCS+RE}, reducing the SMR energy consumption from 30 to 10 kWh_{th}/kg_{H2} gives a marginal improvement of 20% [see Figure S3(b)].

S4.3. Carbon Capture Efficiency

Carbon capture efficiency can be critical in the advancement of the H_{2,SMR+CCS} technology and is shown in Figure S3 (c). Department of energy is looking to increase the carbon capture efficiency from 85% to 99%. Increasing the carbon capture efficiency from 85% to 99% results in a marginal improvement of 14%.

Based on sensitivity analysis it can be concluded that NG leakage and SMR energy consumption are major contributor to net CO₂ emission.

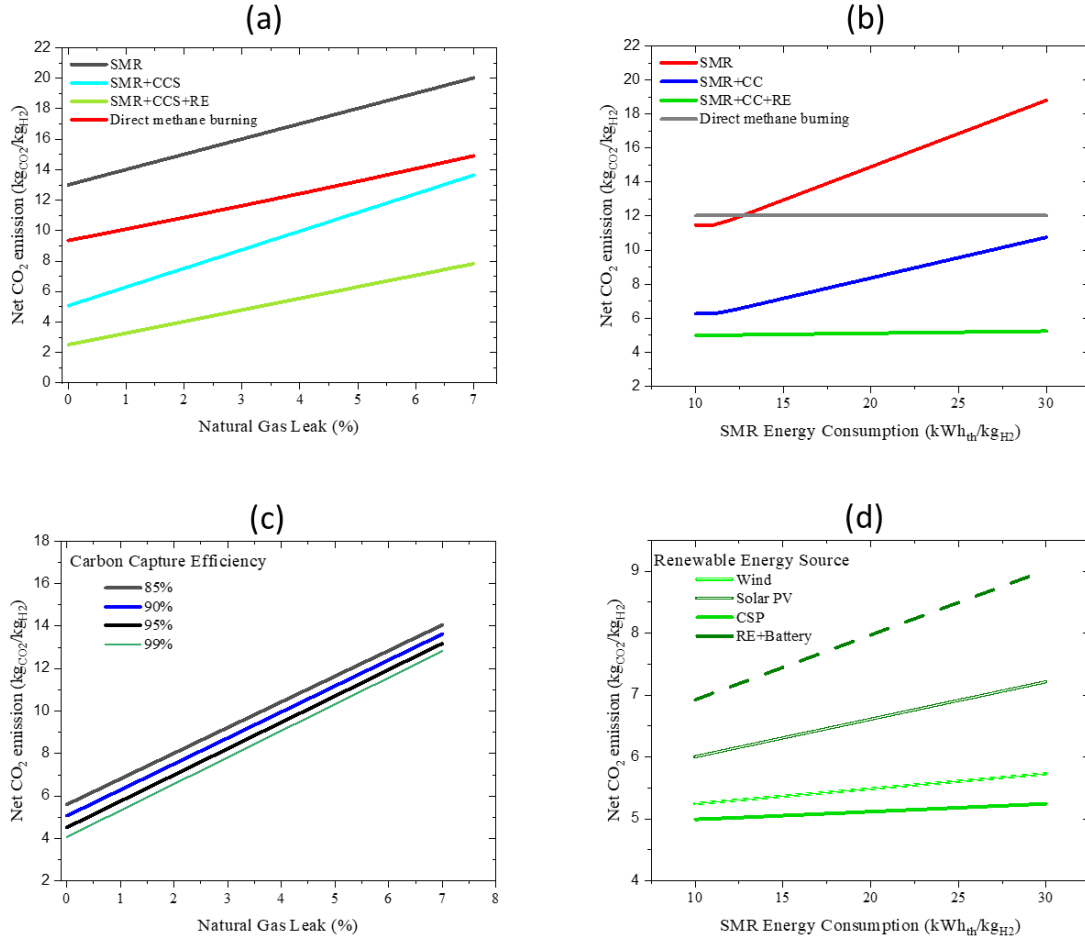


Figure S3. (a) Net CO₂ emission as a function of NG leakage while the SMR energy requirement, carbon capture efficiency and heating energy requirement are kept constant at 24.1 kWh_{th}/kgH₂ and 90%. (b) Net CO₂ emission as a function of SMR energy requirement, while the NG leakage and carbon capture efficiency are kept constant at 3.5% and 90%. (c) Net CO₂ emission as a function of carbon capture efficiency and NG leak, while the SMR heating energy requirement is kept constant at 24.1 kWh_{th}/kgH₂ and (d) Comparison between the performance of H₂,SMR+CCS and H₂,SMR+CCS+RE powered with different renewable energy including solar photovoltaics, wind energy, concentrating solar thermal and battery powered Renewable energy. NG leak, carbon capture efficiency and energy requirement are kept constant at 3.5%, and 90%.

S4.4. Use of Different Renewable Energy Sources as Fuel for Hydrogen

For H₂,SMR+CCS+RE, different renewable energy sources can be used as a heat source including solar photovoltaics (PV), wind energy and concentrating solar thermal (CST). The renewable energy source is an intermittent source of energy and have low annual capacity factor. Where capacity factor is defined as the ratio of actual annual generation to the amount generated had the plant operated at its nameplate capacity for the entire year [61]. Lower capacity factor will reduce the operation hour of the plant. To meet the SMR annual demand, energy storage for renewable energy is required. Since solar PV and wind produces

electricity, we have used battery energy storage. Concentrating solar power typically uses molten storage tank to increase its capacity factor to 60%. The remaining heat can be supplied by $H_{2,SMR+CCS}$.

Figure S3 (d) shows the comparison between different $H_{2,SMR+CCS+RE}$, with respect to $H_{2,SMR+CCS}$. It is interesting to note for $H_{2,SMR+CCS+RE+battery}$, powered by solar PV, the net CO_2 emission is similar to $H_{2,SMR+CCS}$. This is due to lower capacity factor for solar PV system [Table S7], and use of battery storage for the remainder. Battery as an energy storage high carbon footprint and have an efficiency of 80% which further increases the net input electricity required. The overall effect is higher carbon footprint $H_{2,SMR+CCS+RE}$, than $H_{2,SMR+CCS}$. Since wind energy has lower carbon footprint than solar PV and higher capacity factor, using wind energy helps in reducing the CO_2 emission by 40% compared to $H_{2,SMR+CCS}$. CST has the lowest carbon footprint and uses thermal energy storage increases the net capacity factor. This results in significant improvement in the system performance compared to $H_{2,SMR+CCS}$ and can help in reducing the net CO_2 emission by 50%.

Table S9. Operating parameters for different renewable energy sources [60,109,110].

	Capacity factor	Emission (g/kWh _e)
wind	50%	20
Solar photovoltaics	27%	50
Concentrating solar thermal	65%	10
Battery efficiency/emission	85%	100

S5. Approaches to Reduce the Water Consumption for H_2 Production

S5.1. Use of Produced Water Source for Cooling

The cooling water required for electricity generation, SMR and carbon capture are the major contributor to potable water sources (up to 93% of the total water consumption). The cooling tower uses evaporative cooling, where the water gets evaporated. Therefore, the water quality is often limited to potable water source to prevent salt scaling on the cooling tower surface. Instead, once through cooling can be performed where impaired water like produced water (wastewater generated during oil and gas extraction) can be used which can then be sequestered in grade II wells at 100 bar pressure. It may be noted, the produced water generated during oil and gas extraction needs to be sequester. Using produced water for cooling, the net water demand increases to 91, 158, 204 and 156 L/kg_{H₂} for SMR, SMR with carbon capture, SMR with carbon capture powered with renewable energy and regional SMR with carbon capture respectively [see Figure S4]. However, the net potable water demand reduces to 5.7, 5.9, 11.3 and 6.2 L/kg_{H₂} for SMR, SMR with carbon capture, SMR with carbon capture powered with renewable energy and regional SMR with carbon capture respectively. Compared to H_2 generated from electrolyser powered with photovoltaics, the net potable water required reduced by 81%.

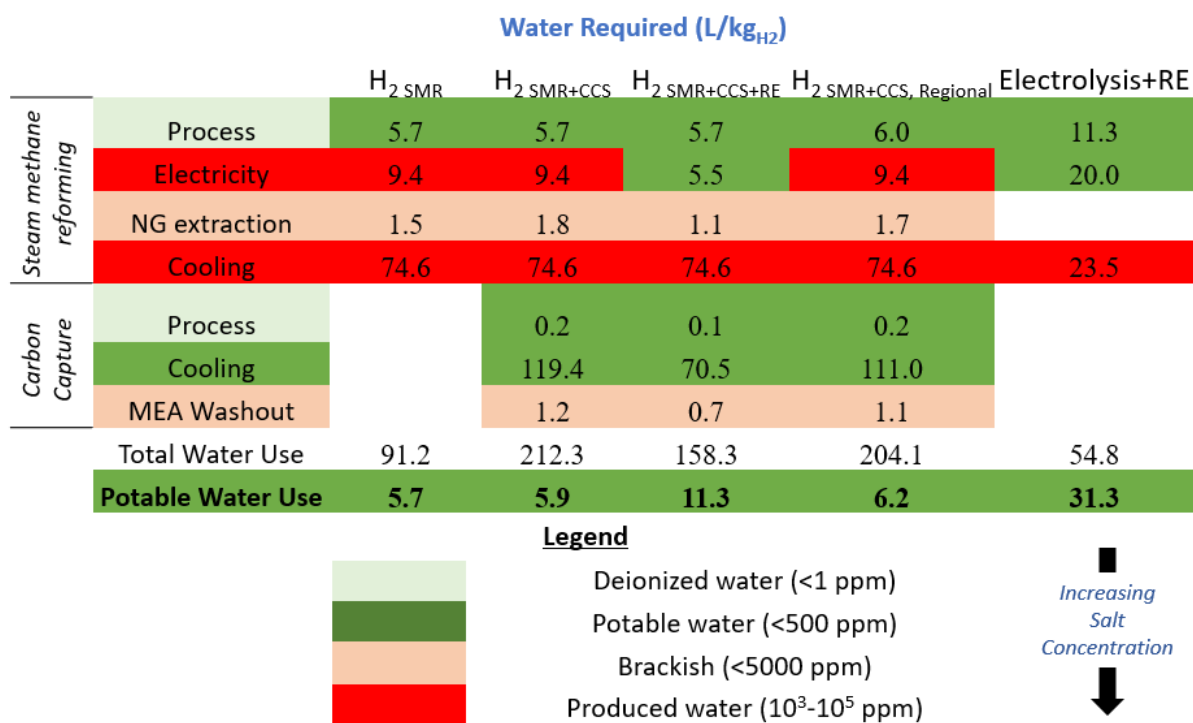


Figure S4. Water requirement for different H₂ production process, with cooling water supplied from produced water.

It may be noted due to significant increase in the produced water required, the pumping power increases. The net effect is the increase in the net CO₂ emission for SMR, SMR with carbon capture, SMR with carbon capture powered with renewable energy and regional SMR with carbon capture by 0.09, 0.27, 0.02, and 0.26 kgCO₂/kgH₂ respectively.

S5.2. Use of Air Cooler for Cooling

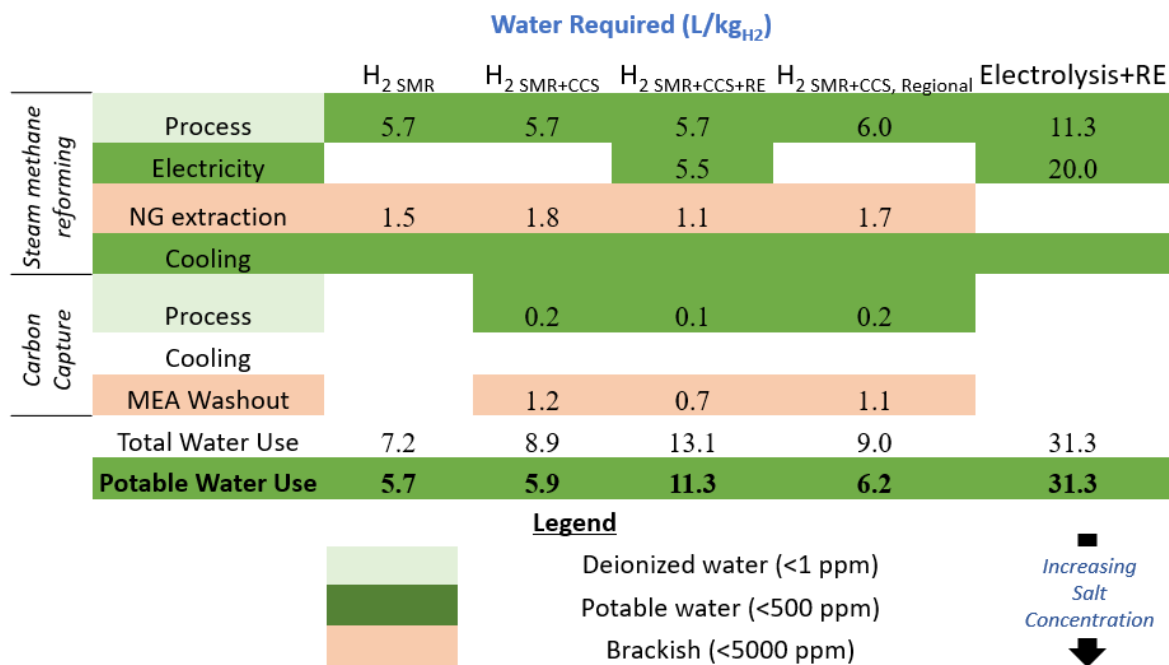


Figure S5. Water requirement for different H₂ production process, with air cooling for electricity, SMR and carbon capture.

The cooling water is the major contributor to net water required, as shown in Figure 7 and Figure S5. To reduce the water requirement, air cooling can be used [63]. This will help in reducing the net cooling water required to electricity generation, carbon capture and cooling of SMR to zero. The total water required reduces to 7.2, 8.9, 13.2 and 9.1 L/kg_{H2} for SMR, SMR with carbon capture, SMR with carbon capture powered with renewable energy and regional SMR with carbon capture respectively. On total water usage basis, the net water consumption for regional H₂ with carbon capture reduces by 71% compared to H₂ generated from electrolysis process powered with photovoltaic. However, with air cooler, the parasitic power consumption increases, thereby the CO₂ production increases by 0.12, 0.27, 0.01 and 0.26 kg_{CO2}/kg_{H2} for SMR, SMR with carbon capture, SMR with carbon capture powered with renewable energy and regional SMR with carbon capture respectively.