Low-carbon and High Purity Hydrogen Production through Sorption Enhanced Steam Methane Reforming (SESMR) with Chemical-looping Combustion (CLC)

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Introduction

- **Hydrogen**, as a versatile energy source, is widely applied in oil refining, chemical production, and iron and steel production, and has also drawn significant attention to tackle various critical energy challenges
- **Steam Methane Reforming (SMR)** is the dominant and commercial technology used for decades for hydrogen production, which is also a large emitter of CO_2- around 2% of the global CO_2 emissions in 2015.
- **Sorption Enhanced Steam Reforming (SESMR)** is an innovative technology to use the pre-combustion CO_2 capture to produce the decarbonised, high purity H_2.
- To reduce the energy penalty and CO_2 emission from the regeneration of CO_2 sorbent in the SESMR, the novel process of SESMR with with **Pressure Swing Adsorption (PSA)** thermally coupled **Chemical-Looping Combustion (CLC)** has been proposed and simulated in Aspen Plus software.

Methodology

Process diagram of the Ca-based SESMR thermally coupled with CLC system

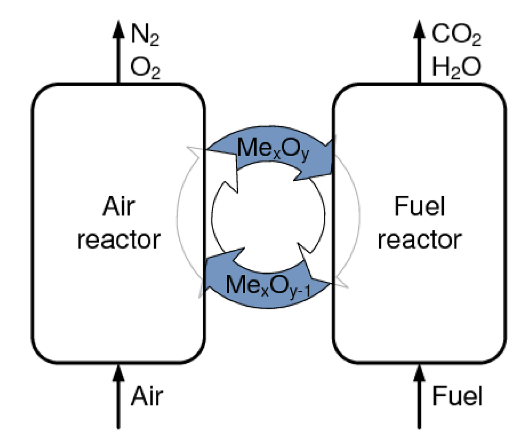
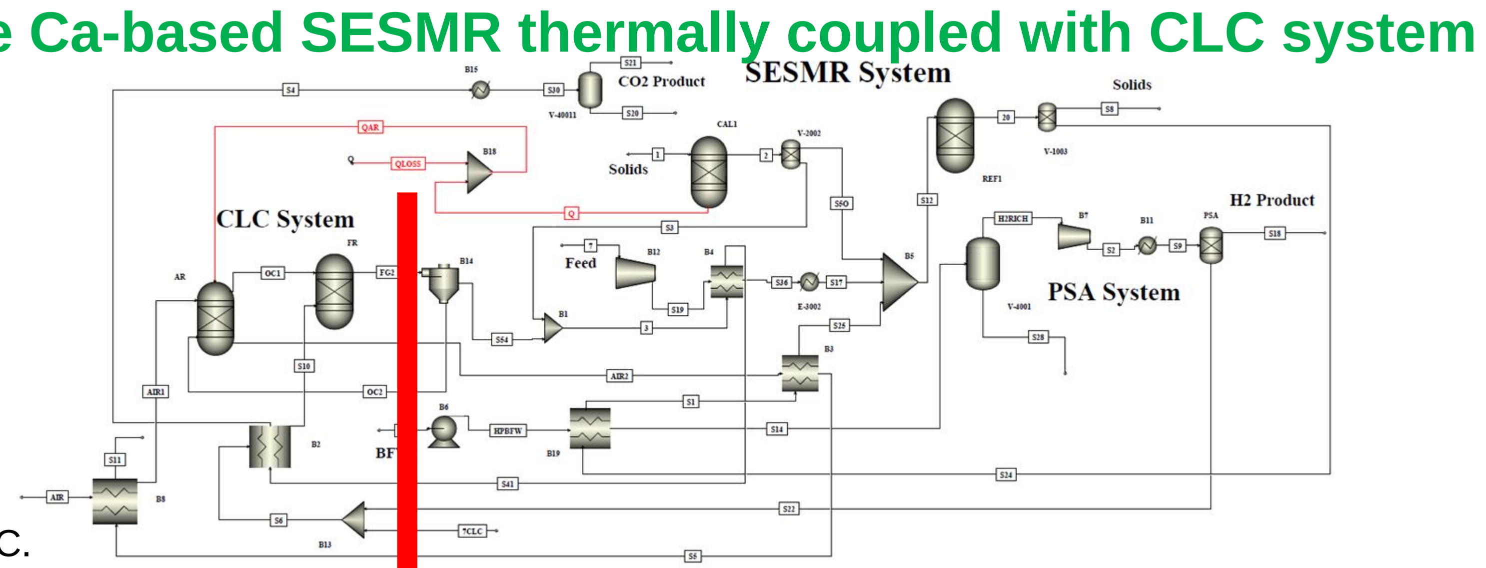
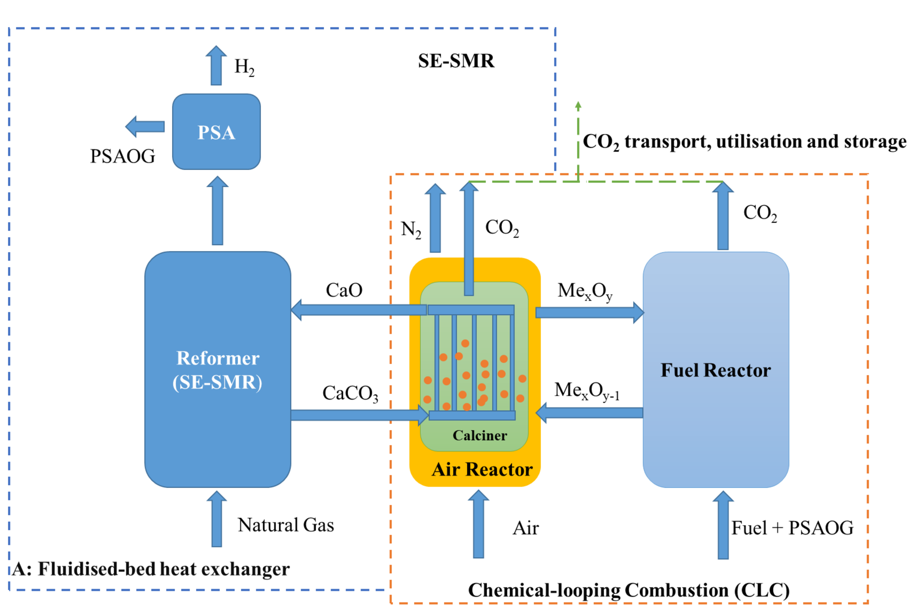


Fig.1 The schematic diagram of CLC.

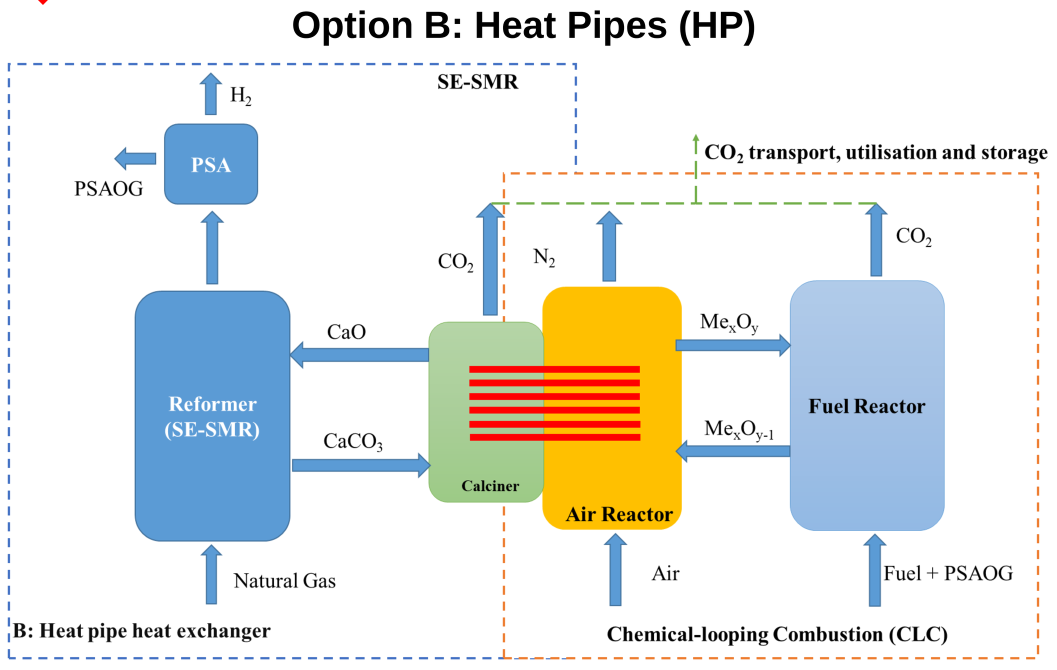


Options for heat integration

Option A: Fluidised Bed Heat Exchanger (FBHE)



Option B: Heat Pipes (HP)



Model development

Reactor	Operating Conditions	Reaction
Reformer	550-850°C, S/C=1-10, Pressure 5-30 bar	$CH_4 + 2H_2O + CaO = 4H_2 + CaCO_3$
Calciner	900°C, Pressure 1 bar	$CaCO_3 = CaO + CO_2$
Fuel Reactor (NiO/Ni)	900-1200°C, Pressure 1 bar, NiO/Fuel =1-7	$CH_4 + 4NiO = CO_2 + 2H_2O + 4Ni$ $CH_4 + NiO = CO + 2H_2 + Ni$ $CO + NiO = CO_2 + Ni$ $H_2 + NiO = H_2O + Ni$
Fuel Reactor (Fe_2O_3/Fe_3O_4)	900-1200°C, Pressure 1 bar, Fe_2O_3/Fuel =3-15	$CH_4 + 12Fe_2O_3 = 8Fe_3O_4 + H_2O$ $CO + 3Fe_2O_3 = 2Fe_3O_4 + CO_2$ $H_2 + 3Fe_2O_3 = 2Fe_3O_4 + H_2O$
Air Reactor (Ni/NiO)	1100°C, Pressure 1 bar, Excess oxygen 5%	$2Ni + O_2 = 2NiO$
Air Reactor (Fe_3O_4/Fe_2O_3)	1100°C, Pressure 1 bar, Excess oxygen 5%	$4Fe_3O_4 + O_2 = 6Fe_2O_3$

Performance Evaluation

- **CH_4 Conversion** = $\left(\frac{n_{CH_4\,in} - n_{CH_4\,out}}{n_{CH_4\,in}}\right)*100\%$
- **H_2 Purity** = $\left(\frac{n_{H_2\,out}}{n_{CH_4\,out} + n_{CO_2\,out} + n_{H_2\,out} + n_{CO_{out}}}\right)*100\%$
- **Cold Gas Efficiency** = $\left(\frac{LHVH_2 * n_{H_2\,out}}{LHVCH_4 * (n_{CH_4\,SESMR} + n_{CH_4\,CLC})}\right)*100\%$

Results

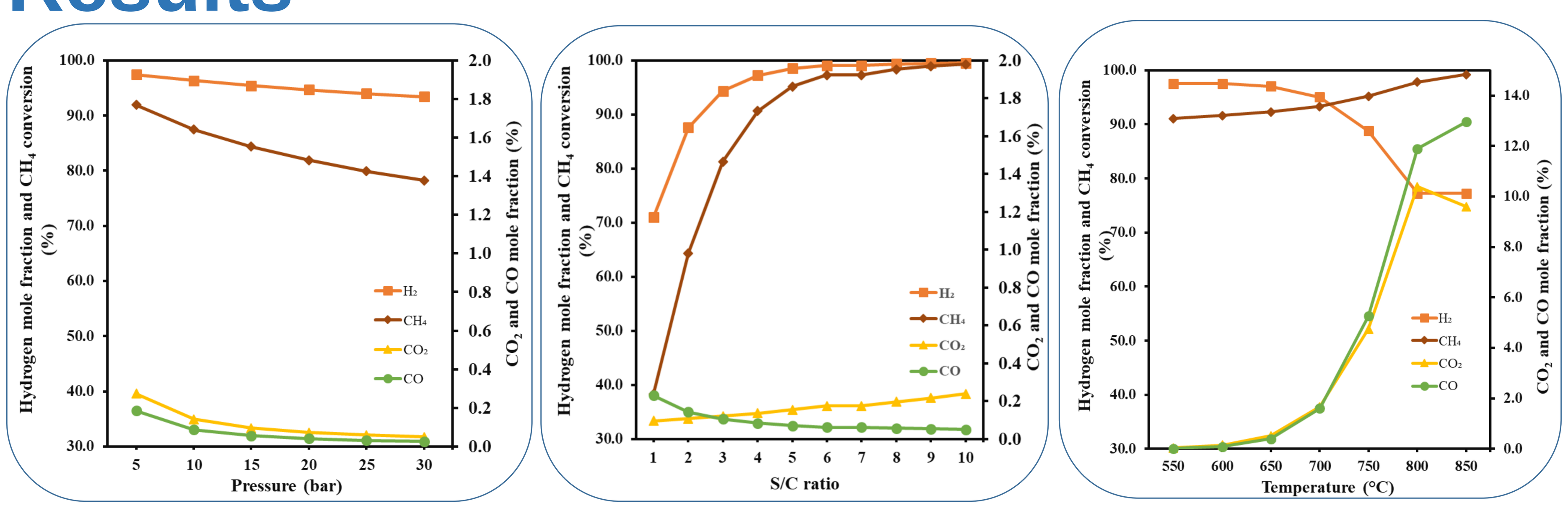


Fig.2 Sensitivity analysis of reformer

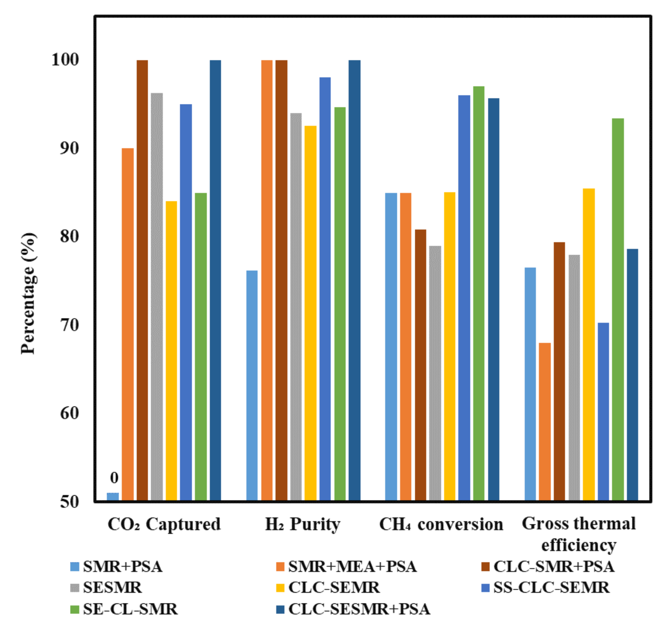


Fig.3 Comparison of different low-carbon SMR technologies

Key Performance Indicator (KPI)	Ni-CLC-SESMR+PSA	Fe-CLC-SESMR+PSA
Reformer: 600°C, S/C=5, 5 bar, CaO/C = 1.9 Fuel reactor: NiO/Fuel = 5 ;Fe_2O_3/Fuel = 9		
CH_4 conversion (Reformer, %)	95.2	95.2
Carbon conversion (FR, %)	99.3	100.0
H_2 conversion (FR,%)	96.7	99.9
H_2 Purity (Dry basis after reformer, %)	98.5	98.5
CO_2 Purity (%)	98.1	97.6
Cold Gas Efficiency (%)	79.1	79.1

Conclusions

- A novel approach of chemical looping concept thermally coupled with **SESMR** combined with **Pressure Swing Adsorption (PSA)** for low carbon and high purity hydrogen production has been presented.
- The iron-based **CLC-SESMR+PSA** process can achieve high conversion of CH_4 (**95.17%**), high gross thermal efficiency (**75.1%**), high hydrogen purity (**>99.99%**) and high overall CO_2 capture (**99.99%**).
- The optimal operating condition for **CLC-SESMR + PSA** according to thermodynamics is at reformer temperature **600°C**, pressure **5 bar**, S/C = **5** and air air reactor temperature 1**100°C**, fuel reactor **900°C**, and molar ratio of Fe_2O_3/Fuel **9**.

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