Dual production of high-purity hydrogen and synthesis gas using integrated sorption-enhanced steam reforming of methane with in-situ CO_2 utilization

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Highlights

- Dual production of H_2 and syngas can be conducted by an intensified thermochemical process.
- High H_2 purity of 89% v/v is produced via sorption-enhanced reforming technology (SE-SR).
- Syngas ($H_2/CO \approx 1.3$-1.5) can be produced by chemical looping bi-reforming (CL-BR) with in-situ carbon dioxide (CO_2) utilization.

1. Introduction

Global major issue regarding to climate change makes H_2 become an interesting substance to fulfill the requirement of searching for alternative cleaner energies. Not only can H_2 be used directly as an energy carrier for transportation but it can also be used as a substance for fuel, electricity, and chemical productions [1-2]. As a consequence, improving the production of H_2 and syngas has attracted great interest as it can not only reduce CO_2 emission but also utilize CO_2 into value-added products. Limiting the emission of CO_2 by introducing carbon capture and utilization unit (CCU) is a choice to improve H_2 production process to be more environmentally friendly and economically. One technique that is employed to improve process efficiency as well as limit CO_2 emission and utilize CO_2 is a co-production of high H_2 purity and syngas using process integration concept [3-5]. In this work, feasibility of the dual production is investigated by studying the effects of steam addition on CH_4 conversion, H_2 purity, syngas quality (the molar ratio of H_2/CO) via process simulation and experiments. Then process design with the consideration of heat exchanger network is proposed.

2. Methods

Thermodynamic and experimental studies for the dual production of H_2 and syngas

Thermodynamic analysis was applied to study the effects of steam to carbon molar ratio (S/C) fed to the calcination reactor on H_2 and syngas production performances and the feasibility of the designed process by Aspen Plus software.

Multi-function material Ni/CaO-$Ca_{12}Al_{14}O_{33}$, composing of Ni as an active catalyst, CaO as an CO_2 sorbent, and alumina as a support material, was synthesized by sol–gel method. The mass fraction of Ni in each catalyst was maintained at 15 wt% with calcium and aluminum ratio 70:30 wt%. H_2 and syngas production tests were experimentally carried out using fixed-bed reactor system.

3. Results and discussion

The results in Fig. 1 show that theoretical CH_4 conversion obtained from the reforming reactor is 90.78%, which is closed to that obtained from the experiments of 85.64%. Simulation result also reveals that high H_2 purity of 76.13 %v/v can be obtained from the reforming reactor, comparative with the experimental result of 68.64 %v/v. The production of high H_2 purity is due to the effect of sorption-enhanced reaction according to Le Chatelier's principle. Syngas can be produced from the calcination reactor; the simulation results show the syngas quality of ca. 1.5-1.6 is obtained for all S/C molar ratios feed whereas the experimental results show the values between 1.2-1.4. The S/C = 0.1 provides the lowest H_2/CO molar ratio and the S/C = 0.5 provides the highest H_2/CO molar ratio. Our results demonstrate that syngas can be produced by chemical looping bi-reforming (CL-BR) with in-situ carbon dioxide (CO_2) utilization.

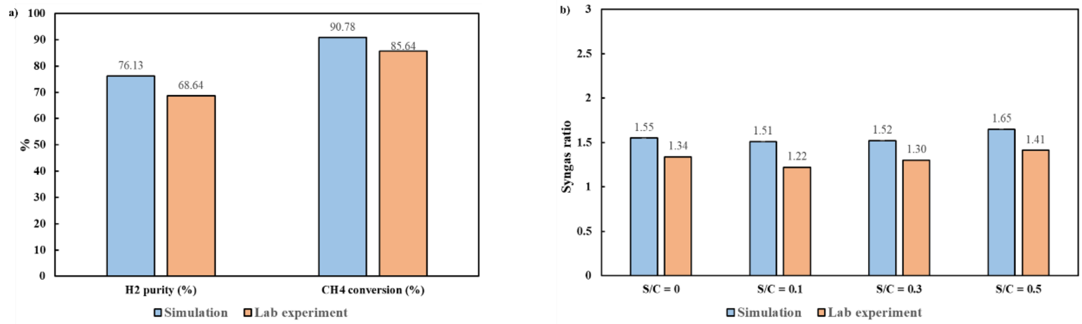

Fig. 1: Comparison of simulation and experimental results a) high H_2 purity obtained from the reforming step and b) syngas obtained from calcination step at different S/C molar ratios.

Process flow diagram of the system is proposed in Fig. 2, the process is composed of three main reactors: reforming, oxidation, and calcination reactors. When calculating and considering the heat exchanger network, the thermal efficiency of the process is found to be 89%.

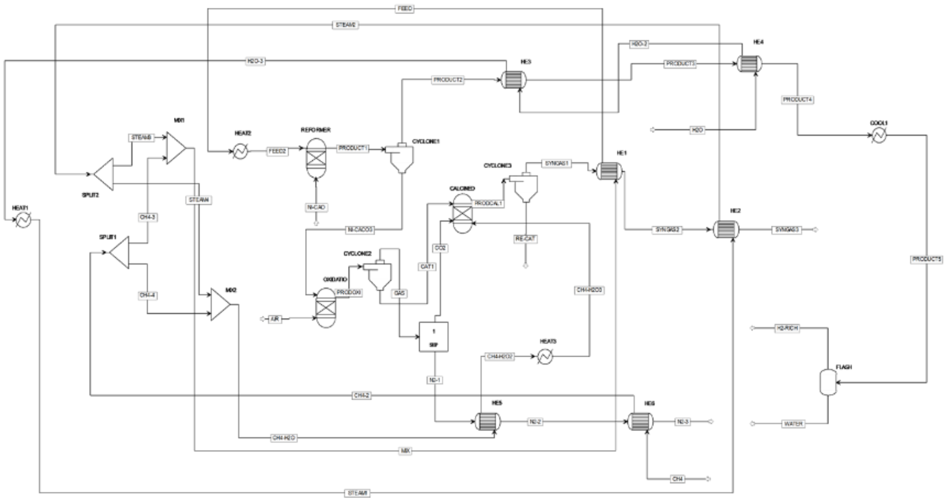

Fig. 2: Proposed process design of an integrated sorption-enhanced steam reforming of methane with in-situ CO_2 utilization for dual production of H_2 and syngas.

4. Conclusions

The results of this study demonstrated that, by utilizing CO_2 under the process intensification concept, high purity of H_2 and syngas could be co-produced. The in-situ utilization of CO_2 to syngas not only can be used as chemical or fuel source but it can improve thermal efficiency of the process.

References

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Keywords

Hydrogen, syngas, CO_2 utilization.