



# Conceptual design of syngas production systems with almost net-zero carbon dioxide emissions



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## ABSTRACT

This work describes three different configurations of syngas production processes using a combination of SMR (steam methane reforming) and DRM (dry reforming of methane). The ideal SMR + DRM process ensures the maximum product yield, the heat-integrated SMR + DRM process fulfills the maximum heat recovery, and the stand-alone SMR + DRM process effectively suppress net CO<sub>2</sub> (carbon dioxide) emissions. Through specific optimization algorithms, the syngas production systems subject to almost net-zero CO<sub>2</sub> emissions are successfully verified by simulations in Aspen Plus environment.

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## 1. Introduction

The efficient syngas production (a mixture of H<sub>2</sub> and CO) is gaining significant attention since it can be used to produce a variety of liquid fuels such as methanol, ethanol, and DME (dimethyl ether) through the Fischer–Tropsch process. SMR (Steam methane reforming), POM (partial oxidation of methane), and DRM (dry (CO<sub>2</sub>) reforming of methane) are the representative reaction processes for syngas production from natural gas. The SMR is a catalytic and energy efficient technology for producing a H<sub>2</sub>-rich syngas [1], but the steam reforming is an endothermic reaction in which the operating temperature is usually over 800 K. The POM is another way to produce syngas where non-catalytic partial oxidation of methane occurs only at very high temperatures between 1300 K and 1600 K and catalytic partial oxidation of methane is a relatively inexpensive alternative option but a significant problem is rapid deactivation due to coke formation [2–4]. The DRM is a potential method in production of CO-rich syngas by consuming greenhouse gases such as CH<sub>4</sub> and CO<sub>2</sub>. This process is attractive from the environmental and economic viewpoint because of the potential utilization of greenhouse gases as resources. Similarly, the main obstacle with respect to the commercialization of the DRM

process is given by the severe catalyst deactivation due to carbon deposition. The highly active catalysts with superior lifetime stability in the DRM have been studied with experimental tests and kinetic analysis [5–7]. The DRM process is not conducive to producing hydrogen since the hydrogen may react with the reactant CO<sub>2</sub> to produce water at specific operating conditions [8]. Through thermodynamic equilibrium analysis of all possible reactions in the DRM, carbon formation as well as water production can be improved by specific ratios of CO<sub>2</sub>:CH<sub>4</sub>:O<sub>2</sub> and modified catalysts at higher operating temperature [9,10].

The SMR is the best option for hydrogen production due to its relatively low cost, but the issues of energy-saving and carbon dioxide reduction have recently been attracting much attention. The heat integration design using heat exchanger network and pinch analysis can be applied to maximize the heat recovery and identify thermal efficiency limit of the SMR process [11–13]. To address the benefits of the combination of different reforming reactions, Supat et al. [14] showed that a combination of non-catalytic partial oxidation and steam reforming has a benefit in terms of balancing the heat load. However, a typical steam reforming with oxygen reactor operated at about 2200 K in the combustion zone and 1200 K–1400 K in the catalytic zone. Song and Pan [15] proposed a novel process with catalytic tri-reforming of methane (SMR, POM and DRM) to achieve high CH<sub>4</sub> conversion and high CO<sub>2</sub> conversion for producing syngas over supported nickel catalysts at 800–850 °C. Halmann and Steinfeld [16] considered flue gases from

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coal, gas, or oil-fired power stations as reactants of tri-reforming of methane to achieve fuel saving and CO<sub>2</sub> emission avoidance. For tri-reforming reactions, the specific catalyst composition and preparation method strongly affect CO<sub>2</sub> and CH<sub>4</sub> conversion. Recently, Zhou et al. [17] showed that the high temperature (over 1173 K) could significantly promote the multi-reforming process while avoiding the problem of catalyst deactivation.

To develop a new process from laboratory scale to industrial scale, the process design, optimization and simulation are critical procedures. Currently, the kinetic models of tri-reforming reactions are incomplete, and the high-activity and durable reforming catalysts are quite rare. In light of kinetic models of DRM and SMR which have been verified by experiments and thermodynamic equilibrium analysis, Gangadharan et al. [18] showed that a combination of SMR and DRM (SMR + DRM) was competitive with the popular SMR. The utility costs and energy consumption of the SMR + DRM process is higher than the SMR process, but this process can effectively reduce carbon emissions. Based on optimal operating conditions, Lim et al. [19] showed that an SMR + DRM process could reduce net CO<sub>2</sub> emission by 67%. However, these SMR + DRM processes need to consume a large amount of external energy to keep the high CO<sub>2</sub> conversion.

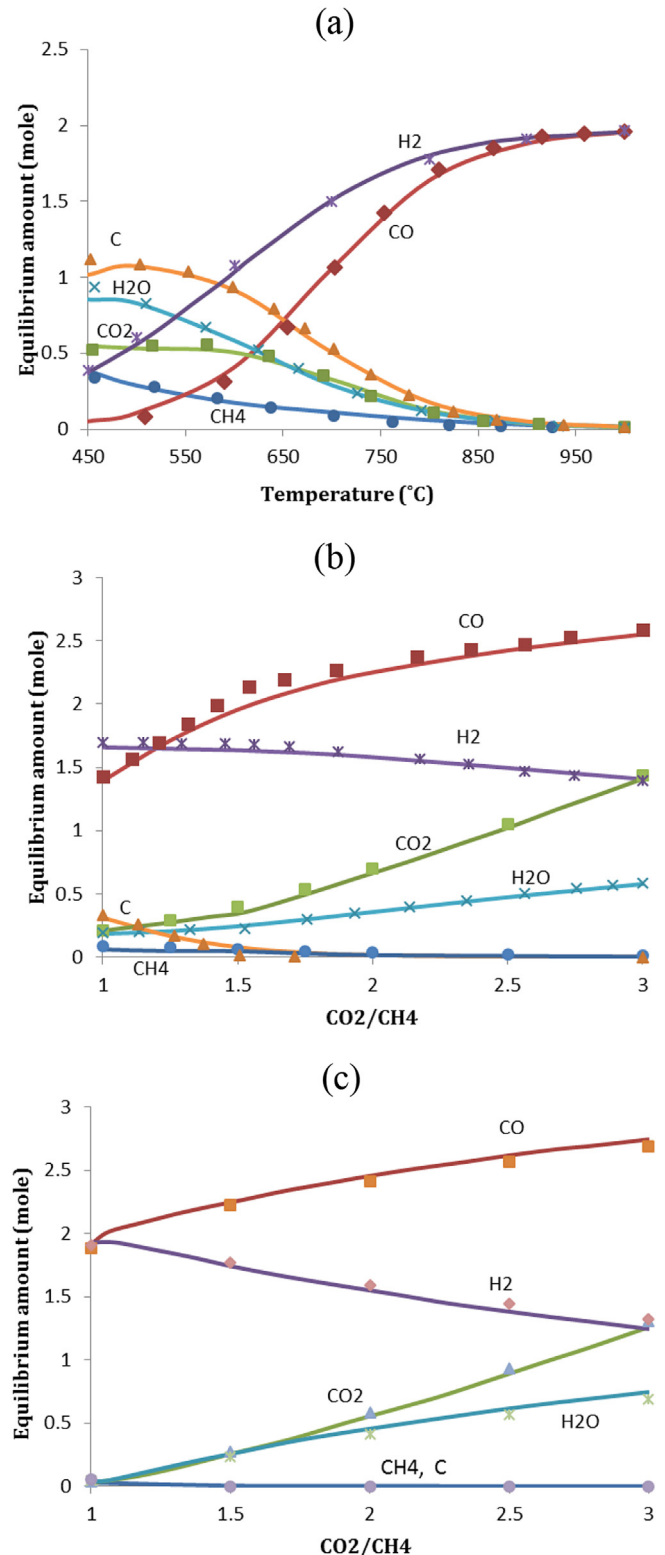
In this article, we propose new syngas production processes to investigate syngas yield, CO<sub>2</sub> emissions and energy consumption. The kinetics and reaction networks for SMR and DRM are introduced in Section 2. Conceptual designs and optimization of three types of syngas production processes are introduced in Section 3. The first design is an ideal SMR + DRM process which can improve the syngas yield and suppress CO<sub>2</sub> emissions. The second design is a heat-integrated SMR + DRM process which can ensure the maximum heat recovery using the heat integration technique. The third design is a stand-alone SMR + DRM process without hot/cold utilities. To address almost net-zero CO<sub>2</sub> emission reduction, those conceptual designs are successfully verified by simulations in Aspen Plus environment.

**Table 1**  
Specifications for process design (I, II, III).

Equipment/Configuration	Aspen module	Specification
SMR Design I&II: Isothermal unit and heater required Design III: Adiabatic reactor	RPLUG	Plug flow reactor: length = 5 m; diameter = 0.5 m; pressure drop = 0.01 atm; catalyst (Ni/MgAl <sub>2</sub> O <sub>4</sub> ) = 0.1 kg
DRM Designs I&II: Isothermal and heater required Design III: Nonisothermal and heating jacket required (UA = 938.2 W/K)	RPLUG	Plug flow reactor: length = 5 m; diameter = 0.5 m; pressure drop = 0.01 atm
VLFD Isothermal	REQUIL	catalyst (Ni/La <sub>2</sub> O <sub>3</sub> ) = 1 kg equilibrium reactor
Pre-combustion (Design III) Isothermal Heat exchanger	Flash2	Two phase flash drum
Design II (EX1, EX2)	RStoic	Stoichiometry reactor
Design III (EX1, EX2, EX3)		Shell and tube exchanger; countercurrent flow Design II: UA <sub>1</sub> = 3398.9 W/K; UA <sub>2</sub> = 2001.2 W/K Design III: UA <sub>1</sub> = 6261.6 W/K, UA <sub>2</sub> = 344.9 W/K, UA <sub>3</sub> = 280.1 W/K

## 2. Syngas production processes

In our study, the SMR and DRM processes are implemented to produce syngas in terms of the ratio of H<sub>2</sub>/CO.



**Fig. 1.** Equilibrium product distribution for the DRM at  $P = 0.1$  MPa vs. (a) temperature at  $\text{CO}_2/\text{CH}_4 = 1$ , (b)  $\text{CO}_2/\text{CH}_4$  at  $750$  °C, (c)  $\text{CO}_2/\text{CH}_4$  at  $900$  °C.

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