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System-Level Performance Analysis of Integrated Thermal Plasma Reformer and SOFC/GT System Using Greenhouse Gases as Fuels

HUAN-LIANG TSAI

Department of Electrical Engineering, Da-Yeh University

168 University Rd., Dacun, Changhwa 51591, Taiwan, R.O.C

ABSTRACT

This paper presents performance analysis of a fuel cell system (FCS) consisting of a thermal plasma reformer (TPR) and a hybrid solid oxide fuel cell and gas turbine (SOFC/GT) system from a system-level viewpoint. This paper also addresses the results of using greenhouse gases (CH_4 and CO_2) as feedstock to the system. Being non-catalytic reforming, the thermal plasma reforming technique does not pose the problems of sulfur poisoning and carbon deposition. With a lower ratio of H_2/CO in the reformate stream, the thermal plasma reforming technique is much preferred to a hybrid SOFC/GT system that exhibits low sensitivity to CO. Through the simulation and analysis of GCtool software, the selected operating condition for the integrated TPR and SOFC/GT system was set at a temperature of 800°C and a CO_2/CH_4 mole flow rate ratio of 1.25. Additionally, the estimated efficiency of the overall system can achieve up to 48% without considering heat loss as a factor. The application of CO_2 reuse contributes to CO fuel production and the reduction of greenhouse emissions from an environmental perspective.

***Key Words*: natural gas dry reforming, thermal plasma reformer, solid oxide fuel cell, gas turbine**

使用溫室氣體爲燃料在整合熱電漿重組器與固態氧化物燃料電池/蒸氣渦輪機的系統層次效能分析

蔡渙良

大葉大學電機工程學系

51591 彰化縣大村鄉學府路 168 號

摘 要

本論文以系統的觀點來分析一個整合熱電漿重組器（thermal plasma reformer, TPR）和混合固態氧化物燃料電池/蒸氣渦輪機（hybrid solid oxide fuel cell and gas turbine, SOFC/GT）的燃料電池系統，運用甲烷、二氧化碳等溫室氣體爲燃料的系統效能。熱電漿重組器本身是非觸媒重組，對於以二氧化碳進行天然氣乾重組不會有硫毒化及碳沈積導致觸媒效能降低的問題；重組混合氣具有較低的氫/一氧化碳比率，適合供應後段的固態氧化物燃料電池/蒸氣渦輪機混合系

統當成燃料。從理論模擬分析，整合熱電漿重組器和混合固態氧化物燃料電池/蒸氣渦輪機的燃料電池系統最佳工作溫度爲 800°C、二氧化碳/甲烷（CO_2/CH_4）進料草耳比値爲 1.25，整體系統效能最高可以達到 48%。從環境保護的觀點來看，二氧化碳再利用來重組甲烷產生一氧化碳與氫氣當成燃料電池的燃料，有助於溫室氣體減量。

關鍵詞：天然氣乾重組，熱電漿重組器，固態氧化物燃料電池，蒸氣渦輪機

I. INTRODUCTION

With increasing concerns about skyrocketing oil prices, global warming, and environmental pollution, the incentives to develop power generation systems with high efficiency and low emission are of great importance. Being the cleanest and the most environment-friendly fuel resource among all primary fossil fuels, natural gas is naturally preferred as the first candidate among available fuels for power generation in the electricity market. On the other hand, fuel cell (FC) with the advantages of low emissions and high efficiency in energy conversion is publicly intended for stationary and mobile power production. Particularly, solid oxide fuel cell (SOFC) system that is a high-efficient of energy conversion and environment-friendly method for electrical power production has been proposed for electric utility power generation in power plants. Together with the effective utilization of the high-temperature exhaust heat in a bottoming cycle leads to a further improvement in the overall efficiency of SOFC system. The integration of SOFC and gas turbine (SOFC/GT) system reaches up to efficiency of 70% [12]. With the improvement of GT technologies and maturity of modular SOFC in recent years, the system efficiency can reach up to 80% [24]. Therefore, the hybrid SOFC/GT is considered to be the most promising technology to achieve the Vision 21 program which was issued by the Unite State Department of Energy (DOE) in 1997 for conceptual feasibility studies fossil power plants with the efficiency higher than 75%. With the increasing availability, high-efficiency hydrogen reforming [2, 4], environmental friendliness, and sufficient infrastructure for refueling, distribution, and storage, the natural gas-fueled, SOFC/GT will play an ever-increasing role in electric power systems in the future.

The principal composition of natural gas is usually methane (CH_4). Traditionally, the catalytic reforming methods of fuel processing system (FPS) are methane steam reforming (MSR), catalytic partial oxidation (CPO), and autothermal reforming (ATR). For the past two decades catalytic reforming of CH_4 with carbon dioxide (CO_2), so-called methane dry reforming (MDR), has been of great growing interest for both industrial applications and environmental friendliness. For industrial applications, the lower ratio of H_2/CO in the reformate stream is suitable for the synthesis of valuable oxygenated derivatives, such as methanol and Fischer-Tropsh syntheses. From a standpoint of environmental friendliness, both CH_4 and CO_2 are known as greenhouse gases (GHG) in abundance in the world. The CH_4 reforming with CO_2 not only enhances the environment-friendly utilization of natural gas but contributes to the GHG-emission reduction. Being more endothermic than MSR process, the MDR process can be used in storing and transporting solar energy [15-16, 27] in the form of chemical fuels to remote areas. A major problem of the MDR reaction is continuous deactivation of catalyst with time, which is mainly due to coke deposition. Many studies have focused on material [12-13, 19-20] or structure [9, 27] of catalyst as well as optimizing the conditions of the catalyst bed [5, 7, 14] to improve the problem of coke formation. In fact, natural gas is a sulfur-containing fuel. Its reformate stream is primarily a mixture of H_2, CO, CO_2, CH_4, and H_2O, and a trace of H_2S. Therefore, the catalytic reforming of CH_4 poses the problems of both sulfur poisoning of catalyst and carbon deposition on the catalytic bed. The non-catalytic thermal plasma reformer (TPR) for the MDR process has proposed to a natural gas-fueled FPS with the characteristics of fuel-flexibility and economical compactness [22]. These contributions motivate us to develop the integrated TPR and SOFC/GT hybrid system for a clean electricity application that has the benefits of low sensitivity to CO, high efficiency conversion, and negligible GHG-emission into consideration.

The main contribution of this paper is to conduct the system-level performance analyses using GCtool software for a SOFC-based FCS taking CH_4 and CO_2 as fuels. The remainder of this paper is organized as follows. For easy presentation, the system description of an integrated TPR and SOFC/GT hybrid system is described in Section II. In addition, the corresponding model is implemented using GCtool package. In Section III, the results of system-level simulation for various operation conditions are illustrated and some discussions are briefed. Finally, brief conclusions are drawn in Section IV.

II. SYSTEM DESCRIPTION AND GCTOOL MODEL

From a system-level point of view, optimization can be performed as a modification of the system configuration. The challenge for an energy system designer is to organize the various components in a FCS system configuration that optimizes the efficiency of FPS, fuel cell, and auxiliaries, utilizes heat to the best extent, and minimizes the heat loss to the external environment. It is costly and time-consuming to obtain an optimal set of system parameters by systematically performing experimental studies. However, the system simulations and analyses can be conducted in order to reduce a number of experiments when many parameters are being investigated in the range of interest. The GCtool (General Computational Toolkit) software package, developed by Argonne National Laboratories [8], allows for several defined inputs to conduct a comprehensive system design and analysis for fuel cell and power generation systems. There were some works [1, 6, 21] mainly using GCtool for investigating the comprehensive performance of a PEMFC-based FCS which consists of a fuel processor, a PEMFC stack, and other supplementary instruments for different configurations and operating conditions. There were only few works of optimizing SOFC-based FCS [3, 17-18] among open literatures. In this study, the system-level performance analysis of an integrated TPR and SOFC/GT power generation system is conducted by GCtool package.

1. System Description

For a system designer, the first stage is to analyze the possible chemical processing requirements and to arrange a system configuration disposing of all the process streams. Fig. 1 shows the integrated TPR and SOFC/GT FCS for the MDR process. The acronyms FC, FM, and TM in this figure stand for flow controller, flow meter, and temperature meter, respectively. The FPS that consists of a reformer, two heat exchangers, and the auxiliaries is to perform a MDR process. The two heat exchangers are used to preheat the CH_4 and CO_2 fuels using the waste heat that is harnessed from the energy of the exhaust gases of gas turbine. A desulfurizer is included in the reformer to remove the sulfur compound H_2S in the reformate stream with a commercial available chemical agent like ZnO. For some sulfur-contained fuels, conventional catalytic reformers utilize a hydro-desulfurizer by injecting hydrogen before entering the reforming reactor. The TPR does not require desulfurizing pretreatment that makes it more compact than conventional ones. Being designed to operate at an atmospheric pressure, the main operating parameters of the TPR are the TPR temperature, inlet temperatures and mass flow rates of natural gas and CO_2.

As shown in Fig. 1, a blower to offer oxygen from ambient air that is preheated by a heat exchanger, the SOFC

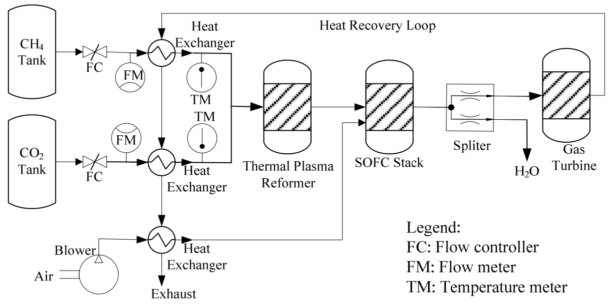

Fig. 1. Schematic diagram of integrated SOFC-based FCS for MDR process.

stack performs the electrochemical conversion, and the GT offers additional electricity for the blower. In addition, a splitter splits H_2O in the gas mixture out of the SOFC's anode. Although pressurized SOFC/GT system has better efficiency up to 70%, an additional SOFC is necessary to be used as a GT combustor. A near atmospheric TPR and SOFC/GT hybrid system is a novel configuration that has not been analyzed in the previous literature. Being operated slightly above atmospheric pressure for a SOFC stack, the high-temperature reformate stream of TPR is directly led to the anode of SOFC stack. On the other hand, ambient air is drawn using a blower, preheated through a heat exchange by recovering the waste heat of the GT exhaust gas, and then inlet into the SOFC's cathode. The chemical reactions occurring inside the SOFC stack that are directly involved in the production of electricity are described as follows [25].

At Anode:

$$H_2+O^{=}\rightarrow H_2O+2e^- \quad (1)$$

$$CO+O^{=}\rightarrow CO_2+2e^- \quad (2)$$

At Cathode:

$$O_2+4e^-\rightarrow 2O^{=} \quad (3)$$

Overall:

$$H_2+CO+O_2\rightarrow H_2O+CO_2 \quad (4)$$

The waste heat of the GT exhaust gas is recovered and harnessed to preheat the inletting fuels of the TPR reactor and the inletting air of the SOFC stack.

2. GCtool Model

Corresponding to Fig. 1, the output diagram of

GCtool-based model for the integrated TPR and SOFC/GT hybrid FCS in is shown in Fig. 2. For easy simulation, the sulfur compound in the natural gas is neglected and CH_4 is taken as a fuel in place of natural gas. Both CH_4 (CH4_Fuel) and CO_2 (CO2_Fuel) fuels are respectively imported from fuel containers to heat exchangers (CH4_HX and CO2_HX) and then directly fed into the TPR (Thermal_Plasma_Reformer). Being operated at atmospheric pressure and high temperature, the reformate stream of reformer is directly fueled into the anode of SOFC stack (SOFC). The ambient air (Air) is drawn by a blower (Blower) and is preheated through a heat exchanger (Air_HX). The preheated air is fueled into the cathode of the SOFC stack. A splitter (H2O_Splitter) is used to separate the un-reacted gas mixture and to product water from the anode. The residual fuels from the anode are harnessed with a GT (Gas_Turbine) to generate an electrical power for the blower. The waste heat of the GT exhaust is harnessed for the three heat exchangers (CH4_HX, CO2_HX, and Air_HX) and the exhaust is finally discharged in ambient air. The waste heat recovery is to makes the system efficiency more attractive.

III. SIMULATION RESULTS AND DISCUSSIONS

As mentioned above, the overall system operates at atmospheric pressure. The main operating parameters of the TPR are the reactor temperature, inlet temperatures of CH_4 and CO_2, and their mass flow rates. The inlet temperature of CH_4 and CO_2 at the feedstock tanks is assumed to be at 25°C. According to the thermal equilibrium prediction of MDR [22], the molar flow ratios of CO_2/CH_4 at 1/1, 1.25/1, 1.5/1, 1.75/1, and 2/1 were analyzed using HSC Chemistry® 5.1 software. For easy analysis in GCtool environment, the molar rate of CH_4 is set as 1 mole/s. The working temperatures of the reformer were set in the range from 500 to 1000°C with an interval of 50°C

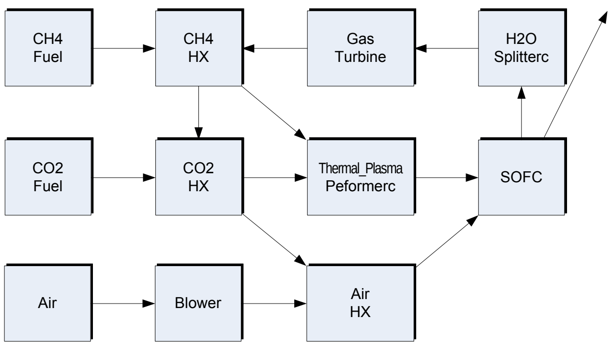

Fig. 2 Output diagram of GCtool for integrated SOFC-based FCS.

increase. The operating temperature and fuel utilization of SOFC stack are chosen at 800°C and 85% [3, 25], respectively. The efficiencies of gas turbine and blower are set at 80%. In this study, the TPR and SOFC/GT are the main power-consumed and power-produced components, respectively. Figs 3-4 show that the TPR power consumption and the total power consumption of overall FCS. The $\dot{M}_{CO_2}$ and $\dot{M}_{CH_4}$ in the figures are molar flow rates of CO_2 and CH_4, respectively. It is obvious that both power consumption increases with the increase of the working temperature of reformer and the increase of the molar flow ratio of CO_2/CH_4. The other power-consuming component is the blower whose power consumption does not increase with a fixed air stoichiometry of 3.0. These results reveal that the higher the operating temperature of reformer is, the more the power consumption of the reformer and overall FCS will be. The increase of $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ means that the amount of CO_2 taking part in MDR reactions increases. The MDR is endothermic that means the power consumption of both reformer and overall FCS will increase. However, this variation makes it possible to increase the total amount of the useful fuels for the successive SOFC/GT hybrid system. The output power of SOFC and the total power production of the overall FCS are depicted in Figs. 5-6. Similarly, both power production increases with the increase of the operating temperature of reformer and the increase of $\dot{M}_{CO_2}/\dot{M}_{CH_4}$. However, both power production does not effectively increase for $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ greater than 1/1 and the reformer temperature greater than 800°C. Being operated at an atmospheric pressure, the total power production of the hybrid SOFC/GT system only depends on the total amount of H_2 and CO fuels. It is assumed that the total amount of H_2 and CO fuels does not significantly increase for the operating conditions that $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ greater than 1/1 and the reformer temperature greater than 800°C. This can be confirmed by the molar fraction analysis of the reformate stream later. Finally, Fig. 7 shows the overall efficiency of integrated thermal plasma reformer and SOFC/GT hybrid system, which the system efficiency is defined as

$$\eta = \frac{P_{\text{out}}}{\Delta \dot{H}_{\text{Fules}} + P_{\text{in}}} \tag{5}$$

where $\Delta \dot{H}_{\text{Fuels}}$ is the variation rate of the total enthalpy of CH_4 and CO_2 fuels, P_{out} and P_{in} are the total power production and power consumption of the integrated thermal plasma reformer and SOFC/GT hybrid system, respectively. The enthalpies of CH_4 and CO_2 fuels are −393.5 kJ/mole and

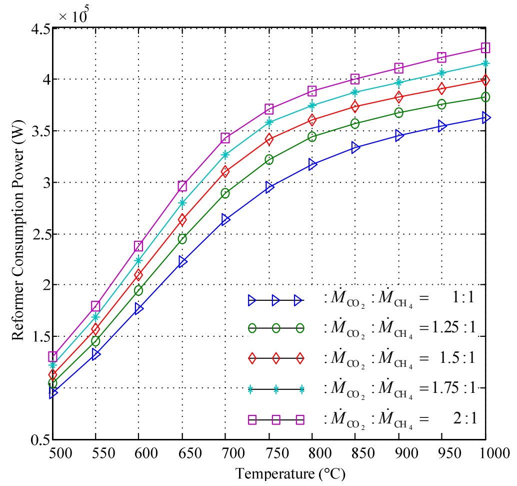

Fig. 3. Power consumption of thermal plasma reformer at different $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ ratios.

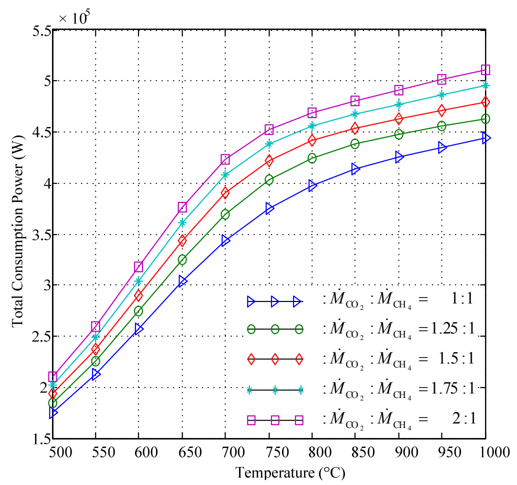

Fig. 4. Total power consumption of overall FCS at different $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ ratios.

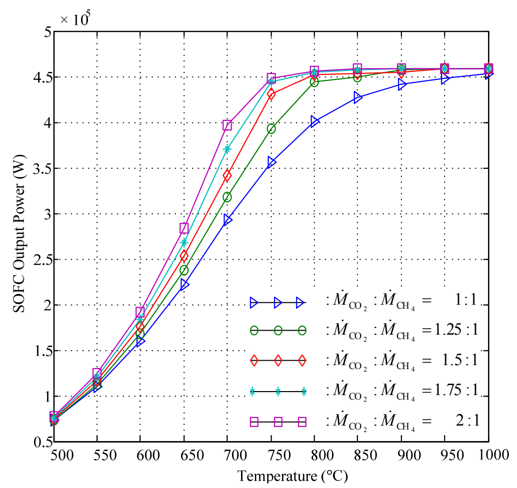

Fig. 5. Output power of SOFC at different $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ ratios.

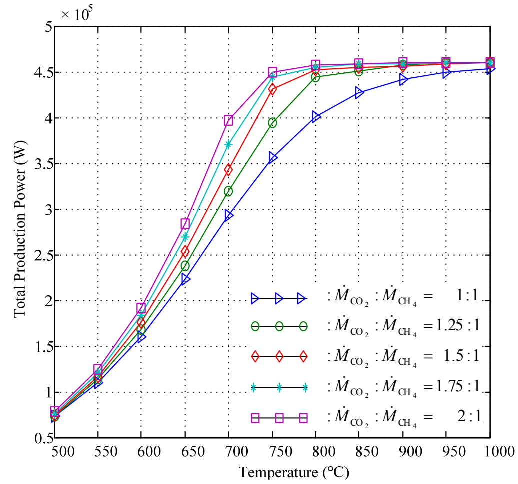

Fig. 6. Total power production of overall FCS at different $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ ratios.

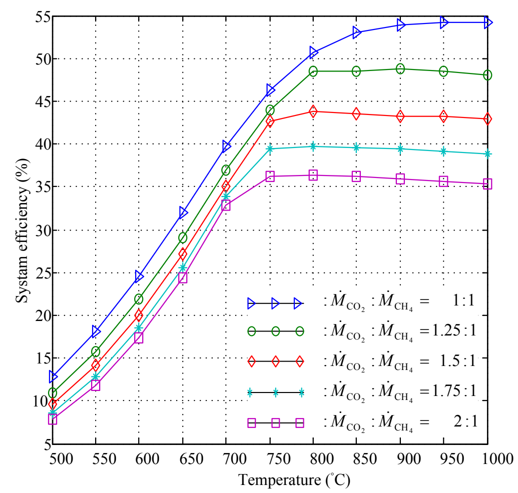

Fig. 7. System-level efficiency of overall FCS at different $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ ratios.

−74.87 kJ/mole at 25°C [23], respectively. At the temperatures lower than 700°C, the reverse Boudouard reaction $C+CO_2 \rightarrow CO$ would be dominant [28]. In order to prevent carbon deposition in the TPR's chamber, the temperature and $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ should be selected at over 750°C and over 1/1, respectively [22]. The optimal system efficiency approaches up to 48% at the selected condition with $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ of 1.25/1 and the reformer temperature over 800°C. On the other hand, the comparison between Figs. 3 and 6 shows that the increase in the power consumption of reformer is higher than one in the power production of useful fuels for SOFC/GT hybrid system with the increase of operating temperature. This means that operating temperature over 800°C does not make the system efficiency increase effectively. The overall efficiency of the integrated TPR and SOFC/GT hybrid system can possibly achieve up to 48% without taking heat loss into consideration. Based on the assumption that both TPR and SOFC/TPR have the design optimization of enclosure insolation, the heat loss of overall system can be assumed to be 5%. Even taking the heat loss into consideration, the overall system efficiency still reach about 45.6%.

From the output file of GCtool for different ratio of CO_2/CH_4 in the range of 500-1000°C, the molar fraction of species in the reformate stream of reformer are shown in Figs. 8-12. GCtool software being designed to deal with liquid and gas fluids, the carbon deposition in component classes is not indicated in the output file of GCtool. Being referred to the thermodynamic equilibrium prediction of MDR process in the TPR [22], the system poses the problem of carbon formation in the chamber at the working temperatures under 800°C and $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ less than 1.25/1. Fig. 8 reveals H_2 molar fractions at different CH_4/CO_2 mole flow ratios. Omitting the case of $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ of 1/1, the best result is that the molar fraction of hydrogen yield reaches over 43% at $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ of 1.25/1 and at temperatures of 850°C. At the same time, the molar fraction of CO approaches approximately 50%. It should be noted that such a lower ratio of H_2/CO is still appropriate for real SOFC operations [10, 28]. Without considering the internal reforming ability of SOFC, both H_2 and CO are the useful fuels directly for SOFC stack. The total mole fractions of SOFC fuels are shown in Fig. 13, which achieve over 94% at temperatures in the range of 700-1000°C for the $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ of 1.25/1. Any higher operating temperature over 800°C can not effectively increase the total molar fractions of SOFC fuels. Higher operating temperature in the reformer leads to higher power consumption and less energy conversion efficiency as well. Therefore, from a standpoint of system efficiency, an appropriate operation condition for the TPR can be chosen at a mole flow ratio of CH_4 to CO_2 of 1:1.25, i.e., mass flow ratio of 16:55, and at a temperature of 800°C for the integrated TPR and SOFC/GT hybrid system.

IV. CONCLUSIONS

System-level performance analysis is a first step to develop a complex fuel cell system. From the system configuration design and system-level performance analysis for CH_4 reforming with CO_2, a selected operating condition of TPR can be with a temperature of 800°C and a mole flow ratio of CH_4 to CO_2 of 1:1.25 (i.e., mass flow ratio of 16:55). With lower ratio of H_2/CO and high conversion efficiency, the reformate synthesized gas is suitable for SOFC/GT-based power generation applications. By integrating with SOFC/GT

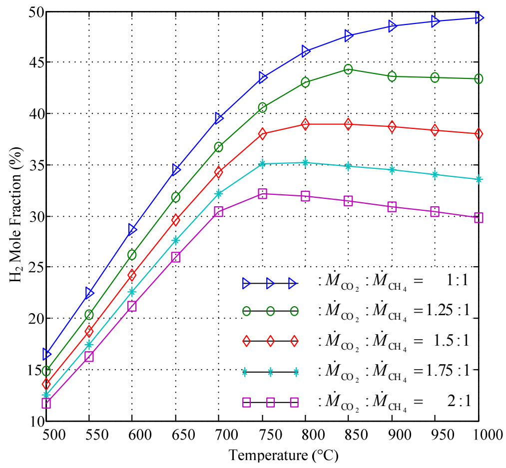

Fig. 8. H_2 molar fractions in reformer output species at different $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ ratios.

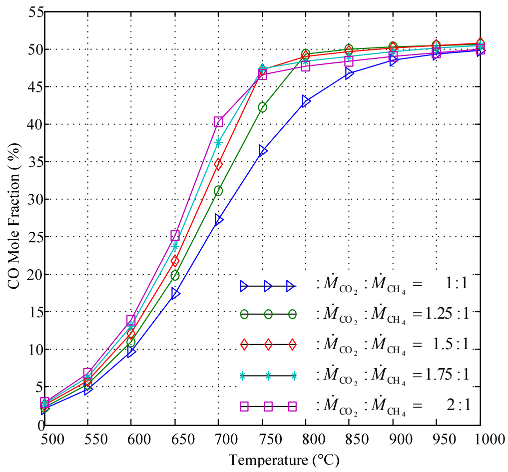

Fig. 9. CO molar fractions in reformer output species at different $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ ratios.

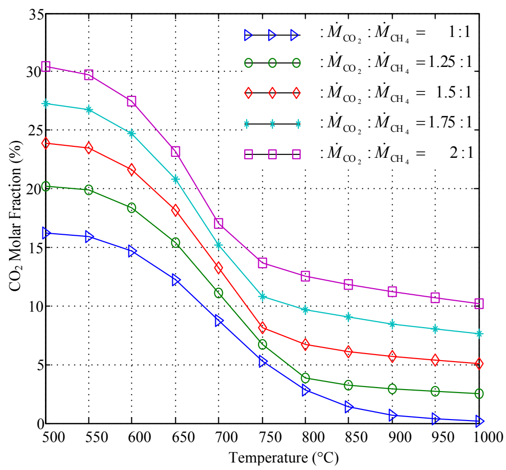

Fig. 10. CO_2 molar fractions in reformer output species at different $\dot{M}_{CO_2}/\dot{M}_{CH_4}$ ratios.

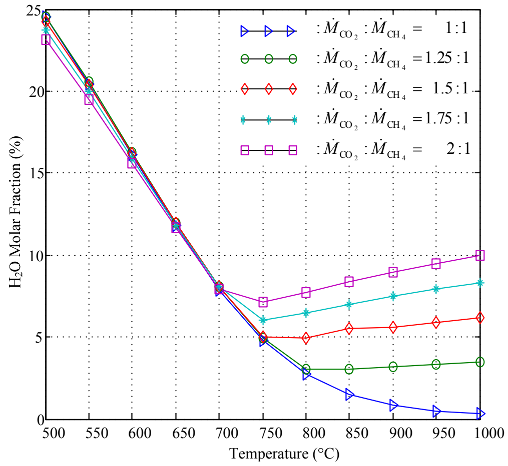

Fig. 11. H_2O molar fractions in reformer output species at different $\dot{M}_{CO_2} / \dot{M}_{CH_4}$ ratios.

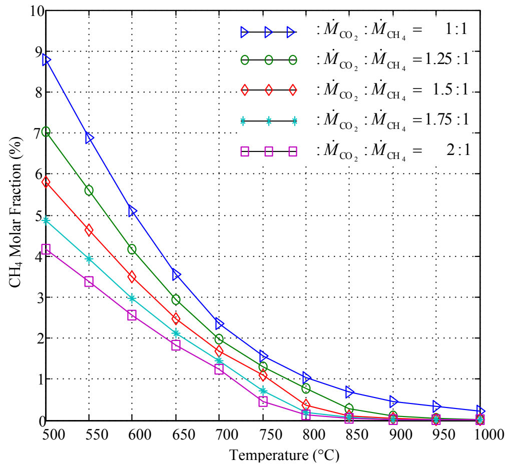

Fig. 12. CH_4 molar fractions in reformer output species at different $\dot{M}_{CO_2} / \dot{M}_{CH_4}$ ratios.

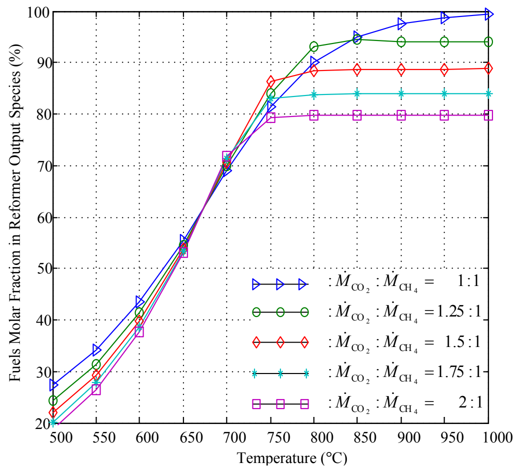

Fig. 13. Total fuels molar fractions in reformer output species at different $\dot{M}_{CO_2} / \dot{M}_{CH_4}$ ratios.

hybrid system, the overall system efficiency can possibly achieve up to 48% without taking heat loss into consideration.

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