Thermodynamic Equilibrium Prediction for JP-8 Steam Reforming in Thermal Plasma Reformer

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ABSTRACT

This paper presents the thermodynamic equilibrium predictions of JP-8 steam reforming in a thermal plasma reformer using HSC Chemistry® 5.1 software from a purely theoretical point of view. The optimal operating condition has been chosen at the temperature of $750°C$ and the steam-carbon (S/C) ratio of 2. The predicted results show that the molar fractions of hydrogen yield in the reformate stream reach 56.25% and 68.86% on wet and dry bases, respectively. For solid oxide fuel cell applications, the molar fractions of total fuels in the reformate stream approach approximately 75% and reach even over 90% on wet and dry bases, respectively.

Keywords: thermodynamic equilibrium, JP-8, steam reforming, thermal plasma reformer

JP-8 燃油在熱電漿重組器進行水蒸氣重組的熱力平衡預測

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摘　　要

本論文從純理論的觀點來分析 JP-8 燃油在熱電漿重組器進行水蒸氣重組的熱力平衡預測。經由 HSC Chemistry® 5.1 軟體模擬分析，可以選定重組器最佳的工作條件：溫度為 $750°C$、水蒸氣與碳(S/C)比值為 2。預測結果顯示重組後的富氫合成氣體中，氫氣的莫耳分量在濕基、乾基條件下分別可達 56.25%、68.86%。對於固態氧化物燃料電池的應用，富氫合成氣體可當成燃料的莫耳分量在濕基、乾基條件下分別可高達 75%、90%。

關鍵詞：熱力平衡預測，JP-8燃油，水蒸氣重組，熱電漿重組器

Ⅰ. INTRODUCTION

According to the report of US Department of Energy (DOE), America is the largest consumer of diesel fuel in worldwide countries. Especially, the diesel consumption of US Air Force and Navy has more than doubled over the last two decades. Therefore, the US military has developed several types of fuels for military use over the past half-century (Maurice et al. [1]). Currently, there are two main types of diesel fuels JP-8 and JP-8+100 used in US military. JP-8 is approximately 99.8% kerosene by weight and additives. JP-8+100 is a version of JP-8 with supplementary additives to enhance its thermal stability. JP-8 is currently used in all US Army and Air Force as well as the aircraft systems of US Navy. However, JP-8 contains three mandatory additives: fuel system icing inhibitor, corrosion inhibitor, and static dissipater. Its combustion emissions, especially polycyclic aromatic hydrocarbon (PAH) compounds, may result in some potential issues of short-term and long-term severe health effects and environmental pollution (Childers et al. [2]; Serdar et al. [3]; Topal et al. [4]; Kellen et al. [5]). In fact, JP-8 has been considered as fuel for fuel-cell-based power supply system for US Army since 1994 (Taschek et al. [6]). A 3MW SOFC-based power plant was demonstrated in 1997 (Abens and Steinfeld [7]). Recently, the researches for the JP-8 fueled SOFC-based power system have been ongoing (Strohm et al. [8]; Gupta et al. [9]; Dreyer et al. [10]; Erri et al. [11]; Roychoudhury et al. [12]). The reformate stream of JP-8 fueled reformer is primarily a mixture of H_2, CO, CO_2, CH_4, and H_2O, and a trace of H_2S as well. However, all of these literatures are focused on catalytic reforming processes which pose the problems of both sulfur poisoning and carbon deposition on catalytic bed. Sulfur compounds must be reduced below 10 ppm. ahead of fuel reformer with a hydrodesulfurizer (HDS) with injecting hydrogen (Strohm et al. [8]; Gupta et al. [9]). This makes both design and implementation of the hydrodesulfurizer somewhat complex. A thermal plasma reformer with a steam reforming process that is a non-catalytic reforming and fuel-flexible method is under progressive development (Tsai and Wang [13]; Tsai et al. [14]). This reforming process combines flameless pyrolysis without combustion, non-equilibrium thermal plasma, and gas-phase superheated steam reforming technologies. Without catalyst being in the reformer, there are no problems of sulfur poisoning and coke deposition on the catalytic bed. After reformation, the sulfur compound H_2S in a H_2-rich reformate stream can easily be removed by commercially available chemical agent like ZnO. These features make the reformer much compact, flexible to operate efficiently on a wide range of hydrocarbon and/or oxygenated hydrocarbon fuels, and better economic competitiveness in the forthcoming energy industry. Since solid oxide fuel cell (SOFC) system can directly use hydrocarbon fuels, the main compounds of JP-8 reformate stream such as H_2, CO, and CH_4 are useful fuels for SOFC-based power systems.

In fact, JP-8 fuel is composed of hundreds of ingredients without a unique chemical specification. In my opinion, large variations of fuel composition in JP-8 will have different thermodynamics for steam reforming using thermal plasma. A limited number of literatures can be found about the thermodynamic analysis of JP-8 steam reforming. Gupta et al. [9] finished a thermodynamic analysis at a steam-carbon (S/C) ratio of 3 and at temperatures in the range of $493-690°C$. In this paper, the HSC Chemistry® 5.1 software package is used to carry out the thermodynamic equilibrium prediction of JP-8 steam reforming. With these predicted working conditions the experiments of JP-8 steam reforming will be conducted in a thermal plasma reformer. The reformate stream can then be analyzed in the hydrogen analysis laboratory in Da-Yeh university immediately. In order to estimate the reformate compositions in a thermal plasma reformer, the thermodynamic equilibrium prediction is first conducted for steam reforming of JP-8 fuel under various operating conditions. These predicted results will be used to compare with the experimental data that are collected through the experiment of JP-8 steam reforming in a thermal plasma reformer. In this paper, the focus is on the prevention of coke formation at an optimal operating temperature with different ratios of JP-8 fuel to water.

The main contribution of this paper is to perform a thermodynamic equilibrium prediction of JP-8 steam reforming in a thermal plasma reformer from a purely

theoretical point of view. For easy presentation, a system description of thermal plasma reformer is firstly introduced in Section II. The thermal equilibrium prediction and some possible chemical reactions of JP-8 steam reforming are then briefed in Section III. In addition, we have some discussions under various operation conditions. Finally, brief conclusions are drawn in Section IV, and future issues of interest are pointed out as well.

Ⅱ. SYSTEM DESCRIPTION

Steam reforming process is an efficient and large-scale process to generate hydrogen in the all ongoing hydrogen reforming methods with hydrocarbon, oxygenated hydrocarbon or H_2-contained fuels and superheated steam at high temperature. Recently, the steam reforming process in a thermal plasma reformer that is fuel-flexible and non-catalytic is under a progressive development (Tsai and Wang [13]; Tsai et al. [14]). Figure 1 shows the schematic block of a thermal plasma reformer at Clean Energy R&D Center, Da-Yeh University in Taiwan, R.O.C. The fuel processing system includes tanks of fuel/water storages, pumps, injectors, atomizer/evaporator, thermal plasma reformer, heat exchanger, replaceable desulfurizer, and electronic control unit (ECU).

Since the thermal plasma reformer can work well under atmospheric pressure without extra pressure, the working pressure of reformer is designed at 1 atm (14.7psi). The temperatures of components, as well as mass flow rates of JP-8 fuel and water are controlled by ECU via the associated control units and switches. JP-8 fuel itself has the properties of high viscosity and low volatility like other fossil fuels. These characteristics sometimes cause the problems of clogging of fuel lines, filters, and injectors. In order to diminish the viscosity value and raise

evaporation rate of diesel fuel, Lee et al. [15] pointed out that with temperature of diesel fuel increasing up to 380 degree in Kelvin (K), its evaporation rate would reach 100%. And the viscosity values of diesel fuel lower under 0.01 (mm^2/s) as temperature increases up to 400K. In order to have low-viscosity and high-evaporation inlet properties, the intake temperature of JP-8 fuel is promoted up to $200\,^\circ C$ in the atomizer/ evaporator. Both JP-8 fuel and water are proportionally injected into the atomizer/evaporator. And the mixture, which is preheated up to $400^\circ C$ by heat exchanger, is then introduced into the chamber of thermal plasma reformer. Thermal plasma hydrogen reformer uses water and H_2-contained fuels to produce hydrogen. It dissociates fuel and water, and utilizes thermal radiation enhancement and high-density insulation to maintain high temperature and increase heat utilization. No catalyst is used; therefore, there is no poison and temperature issues in the reactor. However, there are ions and electrons with high intensity activation inside the reactor's structures, and the reactor has catalyst function. Even for some sulfur-contained fuels, the reformer does not require pretreatment before entering the reactor. After reformation, the sulfur becomes H_2S in a H_2-rich reformate stream, which can easily be removed by commercially available chemical agent like ZnO after the reformer. Heat exchanger is a critical component for the efficiency of transformation that effectively recovers heat energy, efficaciously reduces electricity input, and significantly increases energy and fuel conversion efficiencies. A compact fin tube heat exchanger (FTHE) is adopted to perform the waste heat recovery of the reformate stream. Since JP-8 fuel contains sulfur less than 0.3wt%, a pretreatment of sulfur- contained fuel is required in order to avoid poisoning of catalysts used in the catalytic reformer (Erri et al. [11]), but a desulfurizer is designed for the reformate stream with a replaceable ZnO reductant in the thermal plasma reformer.
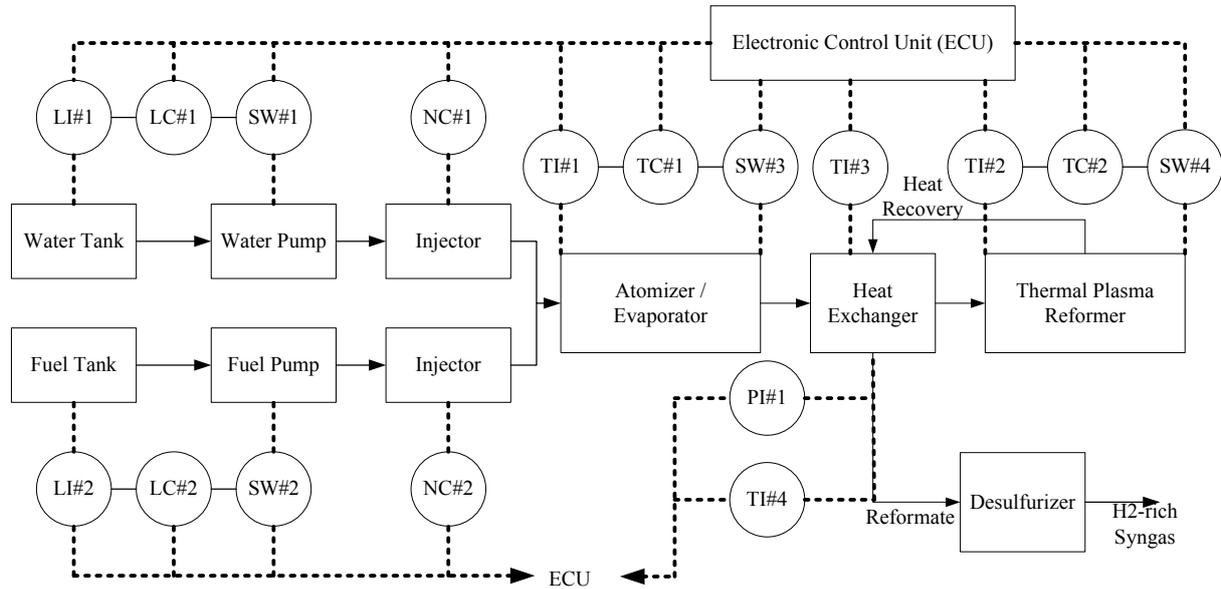
Fig. 1. Schematic block of thermal plasma reformer.

Ⅲ. THERMODYNAMIC ANALYSIS

The thermodynamic equilibrium analysis for the steam reforming of JP-8 fuel using thermal plasma technology is firstly conducted in this paper. In my opinion, the thermodynamic equilibrium prediction should be done ahead to forecast an optimal operating condition and some other working conditions of thermal plasma reformer. The experimental procedure of test plan is then predefined with reference to the predicted results. Without exact knowledge about some possible reactions about steam reforming of JP-8 fuel using thermal plasma reforming technique, I follow the minimization of Gibbs free energy to calculate the thermal equilibrium of JP-8 steam reforming reactions. I adopt a computer code HSC Chemistry® 5.1 to calculate the molar fractions of equilibrium compounds under a given operating condition. The main operating parameters are the temperature and pressure of thermal plasma reformer, inlet temperatures of JP-8 fuel and water, and steam-carbon flow ratio. With JP-8 having uncertain variation of fuel composite, a mixture of JP-8 surrogate typically contains 80.1% alkanes ($C_{13}H_{28}$), 1.9% olefins ($C_{13}H_{26}$), and 18% propyl naphthalene ($C_{13}H_{14}$) as a proxy with a typical mass composition. The equilibrium composition at a given temperature is completely specified with the atomic constituents. The key specifications of JP-8 are

listed in Table 1. A typical mass composition of JP-8 fuel is 85.95% carbon and 14.05% hydrogen. The inlet temperature of JP-8 and steam ahead of the reformer is set at $200\,^{\circ}C$ as mentioned above.

Table 1. JP-8 main specifications

Item	Value	Unit
Composition:		
alkanes ($C_{13}H_{28}$)	80.1	%
olefins ($C_{13}H_{26}$)	1.9	%
propyl naphthalene ($C_{13}H_{14}$)	18	%
Flash point	38(Min)	°C
Viscosity	8.0(Max)	mm²/s, at $-20\,^{\circ}C$
Heating Value	42.8(Min)	MJ/kg

There is an Equilibrium module in HSC Chemistry® 5.1 platform, which enable me calculate the amount of products at equilibrium in an isothermal condition. What I need to do is to specify the amounts and temperatures of the input materials. The equilibrium pressure is the default pressure of 1bar because the thermal plasma reformer is designed to be operated at atmospheric pressure. In addition, the equilibrium temperature is configured at the desired operating temperature in the reformer. The specification can be made in the HSC program

interface. After saving the input data, the final calculation can be conducted. The equilibrium composition is calculated using the Gibbs energy minimization method with the built-in GIBBS solver. The GIBBS program finds the most stable compositions where the Gibbs energy of the reaction system reaches its minimum at a fixed pressure and temperature. The principle of mass balance is a minimization constrain. The thermochemical database of the HSC Chemistry contains enthalpy (H), entropy (S), and heat capacity (C) for more than 17000 chemical compounds and makes the thermodynamic calculation at equilibrium more easily than other methods.

3.1 Thermodynamic equilibrium prediction

I carried out the thermodynamic equilibrium predictions at steam-to-carbon (S/C) flow ratios of 1-5 and at equilibrium temperatures in the range of 500-1000 $°C$ in 50 $°C$ steps. The main compounds of final reformate stream are C (solid residue), H_2, H_2O, CO, CO_2, and CH_4. The corresponding mole fractions on a wet basis are depicted in Figs. 2-7. In the work, there are some considerations taken into account during the thermodynamic equilibrium predictions for steam reforming of JP-8 fuel using thermal plasma: (1) less or free of carbon formation, (2) high selectivity of hydrogen production, and (3) high useful fuels for SOFC applications from a practical point of view. Figure 2 shows that some carbon formations easily take place at lower temperature with S/C ratio of 1. It means that steam is insufficient to react with JP-8 in the form of steam reforming Without enough superheated steam, the superfluous JP-8 fuel directly reacts in the form of thermal decomposition. Therefore, the S/C ratio should be selected at over 1 to prevent carbon deposition in the chamber of thermal plasma reformer. Figure 3 reveals H_2 mole fractions at different mole S/C flow ratios. At S/C flow rate of 2, H_2 mole fractions increase with temperature dependence and approach approximately above 54% on a wet basis at temperature in the range of 700-1000 $°C$. For proton exchange membrane fuel cell (PEMFC) applications, the more the hydrogen selectivity and the less CO concentration of reformate stream has, the better the conversion efficiency

of reformer is. The molar fraction of hydrogen yield reaches 56.25% on a wet basis at S/C flow ratio of 2 and at temperature of 750 $°C$. On the other hand, the total mole fractions of SOFC fuels on a wet basis are shown in Fig. 8. The total mole fractions on a wet basis achieve over 75% at temperature in the range of 700 and 1000 $°C$. The higher operating temperature over 800 $°C$ can not effectively better the total mole fraction of SOFC fuels. The higher the operating temperature is, the higher the power consumption and the less energy conversion efficiency is. Therefore, an optimal operation condition of thermal plasma can be chosen at S/C flow ratio of 2 and at temperature of 750 $°C$ for SOFC application. Figures 3 and 4 exhibit that higher ratios of S/C cause the increase of water and reduction of hydrogen yield with the increase of temperature. It should be noted that the reverse Boudouard reaction $C + CO_2 \rightarrow CO$ would be dominant at the temperatures lower than 700 $°C$ (Wu et al. [16]). This makes, the CO yield increase and the CO_2 decrease with the increasing operating temperature as shown in Figs. 5 and 6, respectively. Figure 7 reveals that methane explicitly decreases with increasing working temperature, which consists with that methane is directly produced by the ethanol decomposition not by the methanation reaction at temperature over 530 $°C$ because the standard free-energy change $\Delta G°$ of methanation reaction becomes positive at temperature of 530 $°C$ (Vasudeva et al. [17]). In order to unify the mass flow rate of JP-8 fuel and water as well as the temperature control design, I choose an optimal operation condition of thermal plasma at S/C flow ratio of 2 and at temperature of 750 $°C$ for both PEMFC and SOFC applications.

Figures 9-14 manifest the molar fractions of C (solid residue), H_2, H_2O, CO, CO_2, CH_4, as well as the total molar fraction of SOFC fuels on a dry basis. The dry basis is to convert the gas concentration on the wet basis into a moisture-free one by neglecting the vapor component. Figure 10 shows the hydrogen yield reaches over 63% on a dry basis at S/C flow ratio of 2 and at temperatures in the range of 600-1000 $°C$. Hydrogen production achieves higher yield on a dry basis with the increase of S/C ratio. This is because that water yield is

getting higher and higher with the increase of S/C ratio as shown in Fig. 4. As shown in Fig. 11, the mole fraction of CO yield on a dry basis increase with the increasing operating temperature. Figures 12 and 13 show the CO_2 and CH_4 yields on a dry basis decreases with the increasing operating temperature, respectively. Fig. 14 reveals that the total mole fractions of SOFC fuels on a dry basis approach over 90% at mole S/C ratio of 2 and at temperature over $750°C$. These all agree with the above analysis on a wet basis. Table 2 presents the mole fractions of H_2, H_2O, CO, CO_2, and CH_4 on both wet and dry bases at $750°C$ with S/C ratio of 2.

Table 2. Molar fractions of H_2, H_2O, CO, CO_2, and CH_4 on both wet and dry bases

Molar fraction (%)	H_2	H_2O	CO	CO_2	CH_4
Wet basis	56.3809	18.3207	17.7048	7.2346	0.3590
Dry basis	68.8607	----	21.4923	9.2074	0.4396
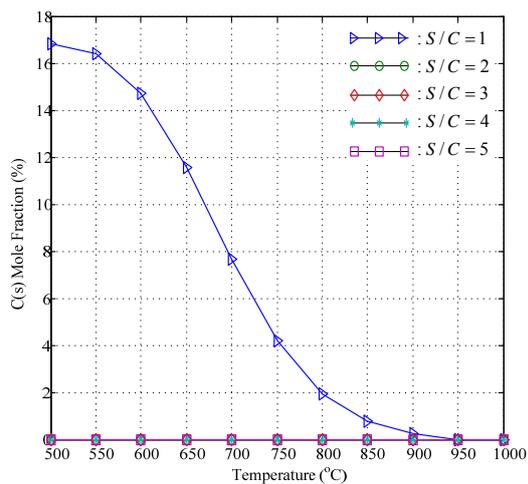
Fig. 2. C(s) mole fractions on a wet basis at different S/C flow ratios.
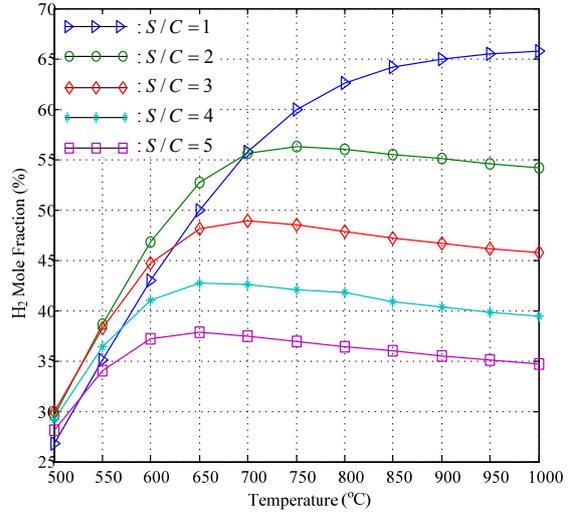
Fig. 3. H_2 mole fractions on a wet basis at different S/C flow ratios.
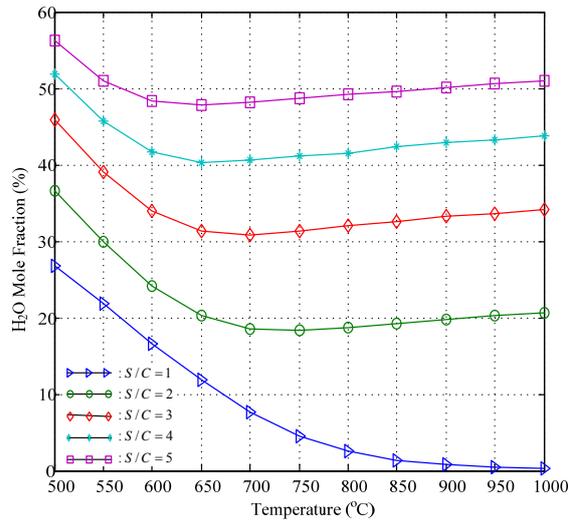
Fig. 4. H_2O mole fractions on a wet basis at different S/C flow ratios.
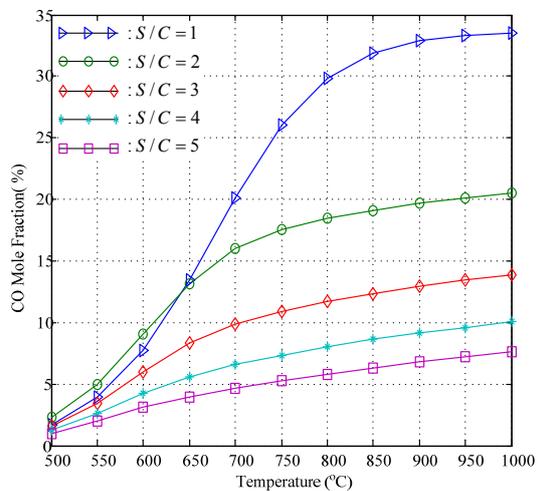
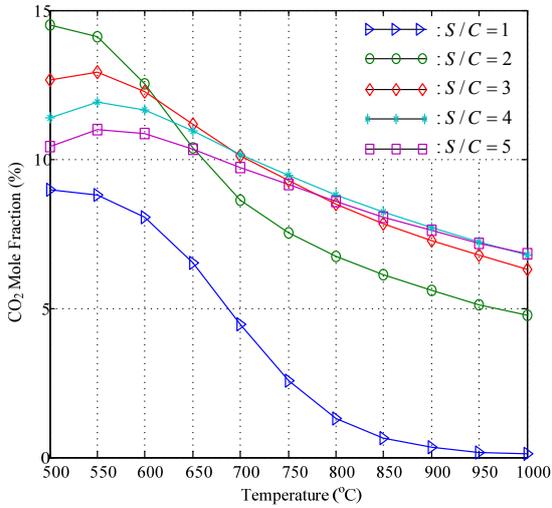Fig. 5. CO mole fractions on a wet basis at different S/C flow ratios.

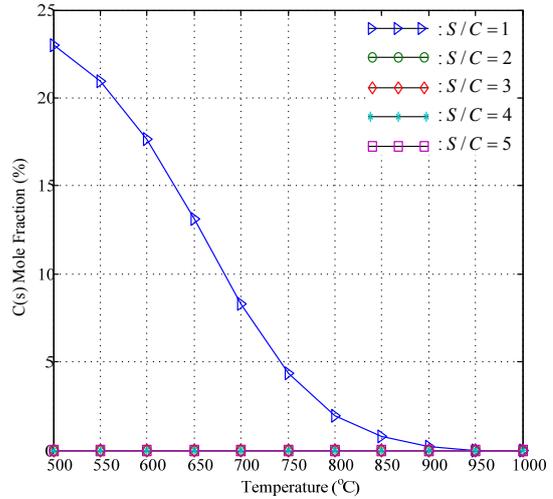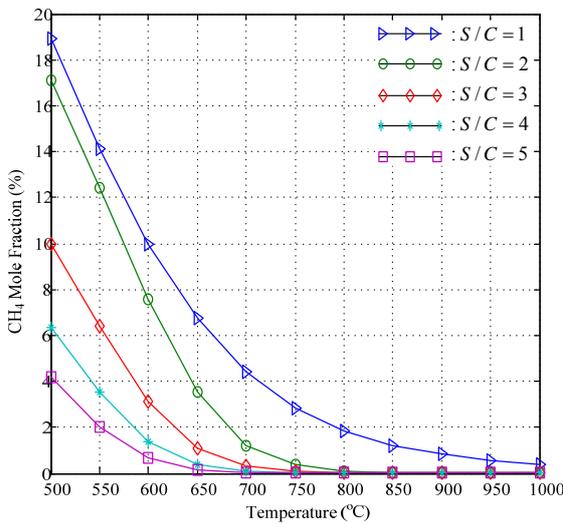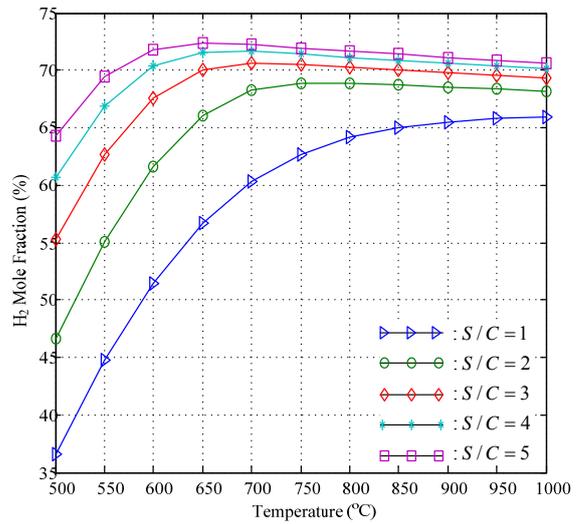Fig. 6. CO_2 mole fractions on a wet basis at different S/C flow ratios.

Fig. 7. CH_4 mole fractions on a wet basis at different S/C flow ratios.
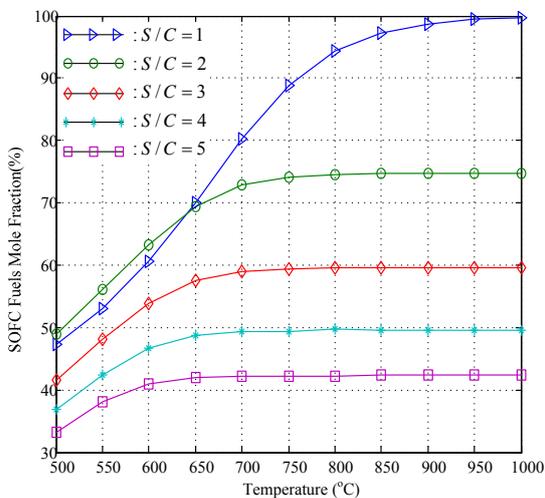

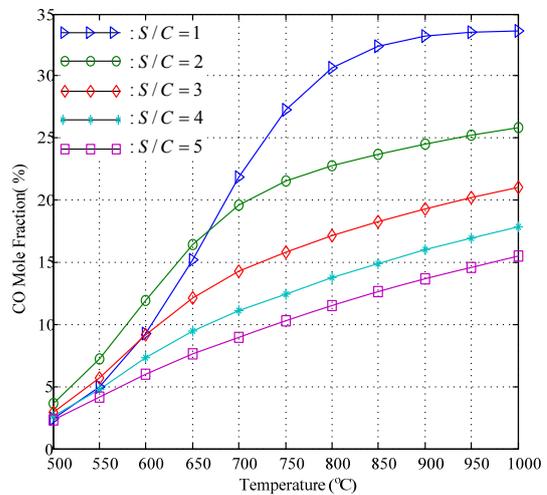Fig. 8. Total mole fractions on a wet basis of fuels at different S/C flow ratios.

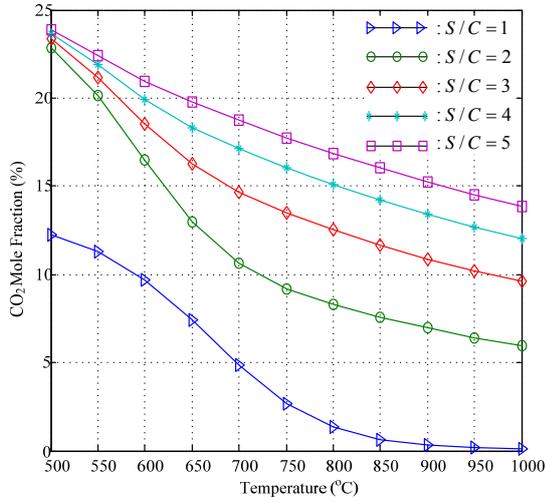Fig. 9. C(s) mole fractions on a dry basis at different S/C flow ratios.

Fig. 10. H_2 mole fractions on a dry basis at different S/C flow ratios.

Fig. 11. CO mole fractions on a dry basis at different S/C flow ratios.

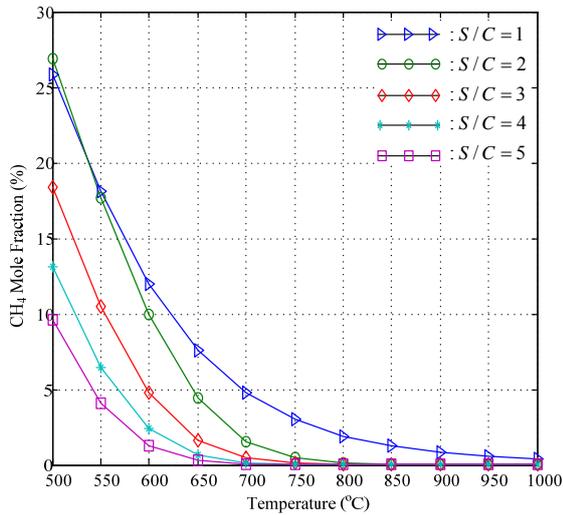Fig. 12. CO_2 mole fractions on a dry basis at different S/C flow ratios.

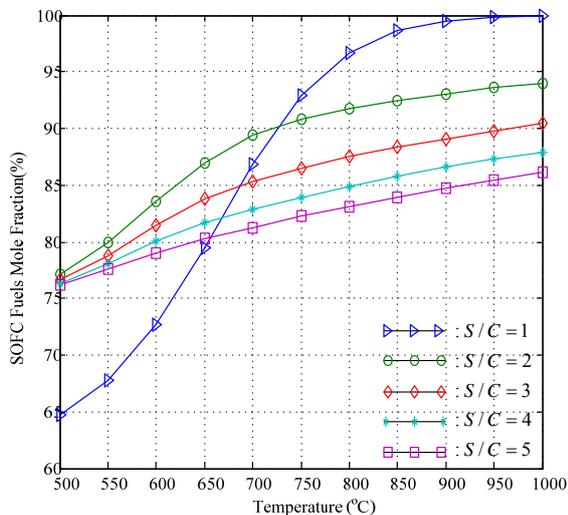Fig. 13. CH_4 mole fractions on a dry basis at different S/C flow ratios.

Fig. 14. Total mole fractions on a dry basis of fuels at different S/C flow ratios.
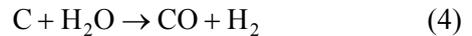
3.2 Possible chemical reactions

JP-8 is basically a kind of kerosene-based hydrocarbon fuel with some additives. With higher energy density and being more environment- friendly than other diesel and gasoline fuels, it gradually becomes a single military fuel for US armed forces. For fuel-cell-based power generation applications, it is necessary to do the thermodynamic analysis of JP-8 reforming. The steam reforming of JP-8 fuel for hydrogen production involves a series of complex chemical reactions. The reactant ratio, operating pressure and temperature are important factors to generate high yield of hydrogen without considerable side reactions and undesired by-products.

Since thermal plasma reforming is a process of flameless pyrolysis, several possible chemical reactions should be considered in steam reforming of JP-8 fuel from a thermodynamic point of view. The thermal plasma reformer is assumed to operate at temperature in the range of 500-1000 °C. Having no knowledge of possible chemical reactions available in the limited number of literatures, I assume that steam reforming of JP-8 using thermal plasma would be modeled as simultaneous pyrolysis and thermal steam reforming of JP-8. To describe the pyrolysis chemistry of JP-8, a pyrolysis-steam reforming mechanism of JP-8 at high temperature is described as follows.
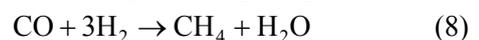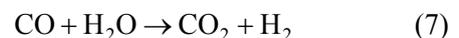
$$2C_{13}H_{25.5} \rightarrow 26C + 25.5H_2 \qquad (3)$$

and

$$C + H_2O \rightarrow CO + H_2 \qquad (4)$$
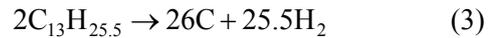
The main reaction of JP-8 steam reforming process is briefly described as

$$2C_{13}H_{25.5} + 26H_2O \rightarrow 26CO + 51.5H_2 \quad (5)$$
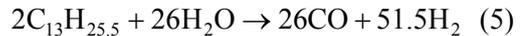
The synthesis gases of reformate stream mainly consist of some gas compositions such as H_2, CO, CO_2, CH_4, and high-temperature steam. The possible chemical reactions taken place in the steam reforming of hydrogen rich synthesis gases are described as follows.

$$CH_4 + H_2O \rightarrow CO + 3H_2 \qquad (6)$$

$$CO + H_2O \rightarrow CO_2 + H_2 \qquad (7)$$

$$CO + 3H_2 \rightarrow CH_4 + H_2O \qquad (8)$$

and

$$2CO \rightarrow C + CO_2 \qquad (9)$$
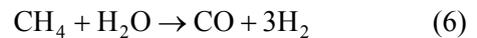
Equations (6) and (7) are called methane steam reforming (MSR) and water gas shift

(WGS) reactions, respectively. These two reactions positively increase the selectivity of hydrogen. Equation (8) is called methanation reaction that produces 1 mole of methane at the expense of 1 mole CO and 3 moles of H_2 consumed. This reaction significantly decreases the selectivity of hydrogen and simultaneously increases the concentration of H_2O production. Fortunately, methanation reaction decreases with the increase of temperature. The coke formation in Eq.(9) is assumed as pure carbon deposition. The analysis of carbon formation is also important, especially necessary for the catalytic reforming applications. It is well know that carbon deposition over the catalytic bed reduces the performance of catalyst and leads to its rapid breakdown. Although there are four possible reactions of carbon formation from a mixture of H_2, CO, CO_2, and CH_4, I have found out carbon formation increases with the increase of CO_2 and the decrease of CO. This carbon formation prediction complies with the Boudouard reaction of carbon formation.

The thermal plasma reformer is assumed to operate at temperature in the range of 500-1000 $^{\circ}C$. It is well known that the standard free-energy change ΔG° of methanation becomes positive at temperature of $530^{\circ}C$. The methanation reaction is significantly suppressed to take place at high temperature of $750^{\circ}C$. The possible reactions including steam reforming and direct thermal decomposition of JP-8 can be postulated in Eqs.(5)-(8). The steam reforming of JP-8 fuel is assumed to be stoichiometrically completed. With the predicted molar fractions of thermodynamic equilibrium at S/C ratio of 2 and at temperature of $750^{\circ}C$, the overall reaction of JP-8 steam reforming in the thermal plasma reformer can be rewritten as

$$2C_{13}H_{25.5} + 52H_2O \rightarrow 57.9361H_2 + 18.8261H_2O$$
$$+18.0883CO + 7.5428CO_2 + 0.3689CH_4 \quad (10)$$
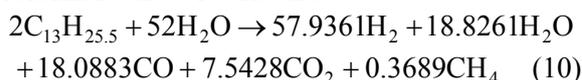
3.3 Discussions

For a sulfur-contained JP-8 fuel, the sulfur becomes H_2S in a H_2-rich synthesis gas after reforming, which can easily be removed by chemical agent like ZnO. For the PEMFC application, the more hydrogen selectivity and the less CO concentration the reformate has, the better the reforming efficiency is. It is necessary

to purify CO concentration as low as possible for the PEMFC system. A High-temperature and low-temperature water gas shift (WGS) reactors as well as preferential oxidation (PROX) reactor are used to purify reformate stream out of CO. With the molar fraction of H_2O being greater than that of CO, the reformate stream can directly introduce into WGS reactors without injecting additional water. This fact makes the CO purification more compact. The schematic diagram of H_2 purification system is shown in Fig. 15. On the other hand, all of the H_2, CO, and CH_4 of reformate stream out of desulfurizer are useful fuels for SOFC stack. The total molar fractions on a wet basis in Fig. 8 approach approximately 75% at temperature in the range of $750-1000^{\circ}C$. The higher operating temperature over $750^{\circ}C$ can not effectively better the total molar fractions of SOFC fuels. Higher operating temperature of reformer leads to higher power consumption and less energy conversion efficiency as well. Therefore, from a control point of view, an optimal operation condition of thermal plasma reformer can be chosen at mole flow ratio of JP-8 to water of 1:26, i.e., mass flow ratio of 181.5:468, and at temperature of $750^{\circ}C$ for both PEMFC and SOFC applications.

IV. CONCLUSIONS

Being more environmentally-friendly and having higher energy density than other diesel fuels, JP-8 will be a potential candidate of single fuel source for all US armed forces. The thermodynamic analysis of JP-8 reforming is a first step to develop a JP-8 fueled fuel cell system (FCS). From above analysis of thermodynamic equilibrium for the steam reforming of JP-8 fuel, I have figured out that an optimal working condition of thermal plasma reformer can be chosen at temperature of $750^{\circ}C$ and mole flow ratio of JP-8 to water of 1:26 (i.e., mass flow ratio of 181.5:468). These predicted results should be verified with the experimental data that will conduct the steam reforming of JP-8 with an existing thermal plasma reformer at Clean Energy R&D Center, Da-Yeh University in Taiwan.
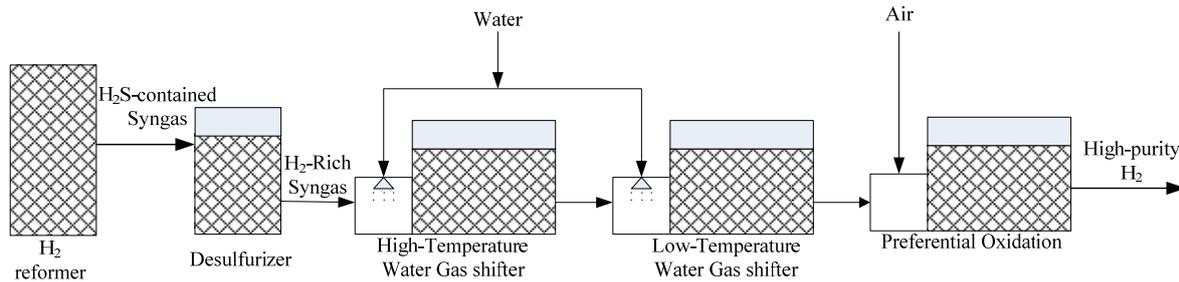
Fig. 15. Schematic diagram of H_2 purification system.

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