Reforming performance of a plasma-catalyst hybrid converter using low carbon fuels

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ABSTRACT

The reforming performance of a plasma-catalyst hybrid converter using different low carbon fuels was investigated. The methodology was to use arc from spark discharge combined with an appropriate oxygen/carbon molar ratio (O_2/C) and feeding rate of the supplied mixture. To enhance the mixing and reforming reaction, a gas intake swirl was generated by inducing the mixture tangentially into the reaction chamber. The required energy for fuel processing was provided by heat released through the oxidation of the air–fuel mixture. The reforming temperature as well as the effect of steam addition on the hydrogen production was studied. The results showed that reformate gas temperature had a profound effect on the overall reaction. The $H_2/(CO + CO_2)$ ratio reformed by both methane and propane was shown to increase with temperature and that the optimum ratio was obtained when reforming methane under 650 °C. The conversion efficiency of the fuel was also shown to increase with increasing temperature. The best thermal efficiency of 72.01% was obtained near 750 °C. The theoretical equilibrium calculations and the experimental results were compared.

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1. Introduction

With increasingly shortage of natural resources and the need for environmental protection, the pressing requirement at the moment is to conserve energy and at the same time look for alternate energy to reduce our dependence on fossil fuel. Current alternate energies include natural gas, methane, ethanol, nuclear and hydrogen, etc., all of which emit less or no carbon dioxide. Of these, hydrogen is recognized as the most promising fuel in future applications, especially as onboard vehicle hydrogen production unit for powering engine or fuel cell. Recently, the use of hydrocarbon fuel reforming to produce hydrogen has been cited as one of the most practical methods in terms of fuel economy and polluting emission reduction for engine applications.

The method of producing hydrogen through plasma or catalytic converters has been studied by various research groups internationally. Bromberg et al. [1] made use of the high temperature and high ionization characteristics of plasma to accelerate chemical reactions during the reforming process. The generation of hydrogen-rich gas from diesel fuel was investigated and they revealed that the evaporation of diesel by preheating provided better process parameters compared with the injection of diesel without preheating. Bromberg et al. [2] revealed that the high energy level

of thermal plasma can accelerate the reforming reaction. In order to reduce energy loss and to improve methane conversion efficiency, they suggested methods of improvement, such as, heat insulation, heat regeneration and improved plasma catalysis. The advantages of such a configuration were the ability of processing a wide range of fuels, high power density, rapid response, low costs, and high fuel conversion efficiency. Horng et al. [3] investigated the effect of intake gas swirl in the fueling process and demonstrated that by enhancing the mixing of fuel and air, the conversion efficiency and the hydrogen concentration can both be improved. They further revealed that high arcing frequency and long residence time of gas in the reaction chamber both resulted in an increasing trend in hydrogen production. Under optimum operating setting, the maximum hydrogen concentration produced was as high as 48% with a hydrogen volume flow rate of 70 mL/min. Futamura et al. [4] studied the reforming of aliphatic hydrocarbon such as methane, propane and neopentane, using non-thermal plasma in the temperature range of 303–433 K. They demonstrated that reforming by combining carbon dioxide with methane and propane, yielded a higher H_2/CO ratio than using neopentane. When using steam reforming, however, methane yielded the highest H_2/CO ratio.

Ayabe et al. [5] investigated autothermal reforming of methane and propane by 2 wt.%-catalysts on alumina support. They proposed the activity level of the catalysts in descending order as: Rh > Pd > Ni > Pt > Co. They demonstrated that high activity was

achieved by loading suitable amounts of Ni, while under the same loading, activity was higher for the Rh-based catalysts. Ma et al. [6] experimented with different fuels on the light-off effect of catalyst and compared the performance of autothermal reforming of methane, ethane and propane with a Pt/δ-Al$_2$O$_3$ catalyst. The experiments showed that a Pt catalyst resulted in better conversion efficiency compared to a Ni catalyst and that under low temperature, the reforming efficiency of propane was found to be better than ethane and methane. Cipitì et al. [7] developed a system for fuel cell application by converting low carbon fuel into hydrogen through autothermal reforming in a Pt/CeO$_2$ catalyst. Commercialized catalysts were then employed for intermediate water gas shifting (ITWS) and preferential oxidation (PROX) at the rear part of the system under an operating temperature of 600 °C, 320 °C and 120 °C, with the respective light-off temperature of 300 °C, 260 °C and 90 °C. Preliminary results showed the need of improving the prototype engineering and they reported that additional tests were in progress to verify the performance. Kantak et al. [8] studied the reforming of low carbon fuels such as natural gas and LPG with an S/C ratio of approximately 0.5 and an O$_2$/C ratio of 0.4–0.7. The fuel conversion efficiency of natural gas was found to be as high as 92.9% and the thermal efficiency was 76.9% with a H$_2$/CO ratio of about 2.13. In comparison, LPG yielded a maximum fuel conversion efficiency of 88.5%, a thermal efficiency of 72.5% and a H$_2$/CO ratio of 1.44. Using an Rh/Al$_2$O$_3$ catalyst to reform propane, Silberova et al. [9] obtained an optimum C/O ratio of 0.8 at a temperature of 300–1000 °C. Under partial oxidation at a furnace temperature of 700 °C, optimum hydrogen production was attained. At 800 °C, the conversion efficiency of propane was as high as 100%. Horng et al. [10] designed a small scale plasma converter for application on a four stroke motorcycle. They showed that by adding heat storing material at the exhaust pipe, the exhaust emission was reduced during cold start.

Due to its improvements to hydrogen production and savings in plasma power, the plasma-catalyst hybrid unit has been increasingly employed. Bromberg et al. [11] suggested that the combination of partial oxidation with a suitable catalyst could reduce CO concentration and improve hydrogen yield, thereby reducing plasma power consumption. Bromberg et al. [12] combined the hydrogen-rich gas produced by a plasma converter with EGR and then by feeding the mixture into an internal combustion engine, they demonstrated that the NO$_x$ emission could be effectively reduced as would the HC emission during cold start. Sobacchi et al. [13] studied the characteristics of hydrogen produced from isooctane and reported that hydrogen yield at catalyst temperatures as high as 800 °C was significantly improved by using a combined plasma/catalytic system. Pietruszka et al. [14] employed a plasma catalytic reactor for the combined oxygen-steam reforming of methane. They found that by combining DBD and a Ni catalyst the conversion of methane was not improved, but full oxygen conversion was achieved. Under a suitable temperature to maintain the Ni catalytically active, the product selectivities changed significantly. Petitpas et al. [15] reviewed available literature on the field of assisted production of hydrogen by non-thermal plasma reforming. They concluded that, thus far, most of the existing reforming reactors were still being developed and advanced in research laboratories.

In light of the above, the increasingly stringent regulations on emission and the shortage of energy, the application of reformers on vehicles to produce hydrogen could be one of the future directions of internal combustion engines and fuel cells. In this study, a plasma-catalyst hybrid system with emphasis on motorcycle application was designed and investigated. Low carbon fuels, such as methane and propane were used for hydrogen-rich gas production, allowing, therefore, the results to be easily applied to natural gas or LPG reforming. The system has been installed on a motorcycle and currently in tests. It is anticipated that the results would soon be published.

2. Test set-up and methods

The main apparatus was a plasma converter that was fueled with low carbon fuel for producing hydrogen. The experimental set-up is shown schematically in Fig. 1. The reformer included three sub-systems: the fuel supply system, the reforming unit and the emission sampling system. The fuel supply system included nozzles and flowmeters for regulating the ratio of the mixture. With the aid of the purpose-designed reaction chamber, intake swirl was produced to promote mixing of the fuel and air. The reforming unit consisted of a spark generating system and a catalytic converter. The former was modified from a car ignition system for compatibility of the current set-up for use in a vehicle
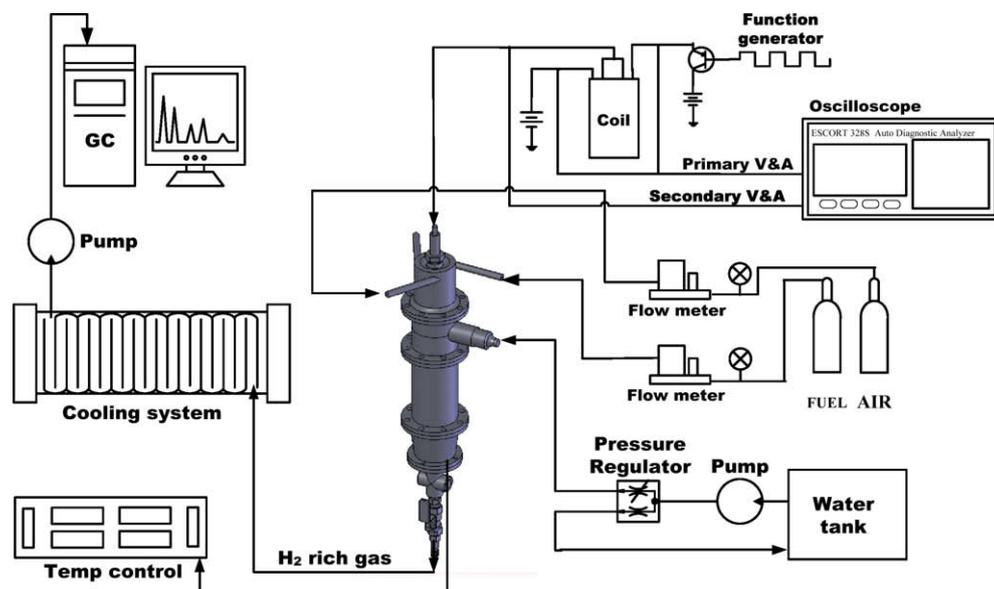

Fig. 1. Schematic of the experimental set-up.

Table 1
Specifications of the reformer.

	Primary side	Secondary side
(a) Power supply unit		
Voltage (V)	12	4000
Current (A)	3.0	0.004
Power (W)	36	16
Frequency (Hz)	200	200
(b) Catalytic converter		
$D \times L$ (mm $\times$ mm)		$\Phi 46.2 \times 50.0$
Pt/Rh weight ratio		5/1
Loading(g/ft^3)		50
CPSI (cell/in^2)		100

in order to reduce cost and space. The specifications of the system are shown in Table 1a. Spark arcing at the electrodes was produced by generating a high voltage at the coil through the signal generator. The anode was made of an NGK BP5ES spark plug with the ground electrode removed. The cathode was the main body of the reaction chamber. The specifications of the commercialized catalyst are shown in Table 1b. The catalyst was chosen mainly for its low cost and availability, as well as its low pressure drop and high mechanical strength. Moreover, its metallic substrate has the advantage of a quick cold start regardless of using liquid or gaseous fuel. It is thought that the above advantages can be directly applied to internal combustion engine or fuel cell with the advantage of low development costs.

In terms of temperature measurements of the gas mixture, three K-type thermocouples were positioned at the inlet, mid-section and outlet of the catalyst. The average of these three measurements was recorded as the reformate gas temperature for comparison with theoretical equilibrium analysis. The produced gas was collected by a gas sampling bag for analysis by an Agilent 6850 gas chromatographer. The commercialized HSC Chemistry software (©ChemSW Software, Inc.) was used to calculate the equilibrium production.

There are currently a few hydrogen production processes in the industry. In this study, the method of reforming was adopted.

Reforming processes include partial oxidation (POX) reforming, autothermal reforming (ATR), steam reforming (SR), catalytic decomposition and coal gasification, etc. The mechanisms of the various reforming processes are summarised in Fig. 2. This study used low carbon fuels, namely, methane and propane for producing hydrogen. Partial oxidation and oxidative steam reforming (OSR) were applied. Where, oxidative steam reforming is the reaction of partial oxidation combined with steam addition. The relevant operating parameters are shown in Table 2. Methane and propane were supplied in gas tanks whilst air was fed by an air compressor. Both were separately directed into the plasma reactor via nozzles using the tangential induction method. For the partial oxidation process, an appropriate amount of oxygen was input to provide sufficient oxidation heat for reforming in the catalytic converter. The hydrocarbon fuel was reformed into hydrogen and carbon monoxide. Further reforming was carried out by adding an appropriate amount of steam in order to enhance hydrogen production. Eq. (1) expresses the theoretical reaction and Eq. (2) the actual reaction.

$$C_nH_mO_p + z_0(O_2 + 3.76N_2) + (2n - 2z_0 - p)H_2O \rightarrow nCO_2$$
$$+ (2n - 2z_0 - p + m/2)H_2 + 3.76z_0N_2 \qquad (1)$$
$$C_nH_mO_p + z(O_2 + 3.76N_2) + yH_2O \rightarrow x(aCO + bCO_2$$
$$+ dH_2 + eO_2 + fCH_4 + gN_2) + hH_2O + jC \qquad (2)$$

where z_0 is the coefficient of the theoretical reaction, z, a, b, d, e, f, g, h and j are those of the actual reaction and x is the ratio factor.

The reaction range of partial oxidation is very wide, reforming in the range between cracking and complete oxidation. Fig. 3 shows the comparison of enthalpy of reaction in the reforming process under different O_2/C ratios for methane and propane. The different letters in the figure represent each type of reaction process for each type of fuel. It is noted that an appropriate level of O_2/C ratio is important. For low O_2/C ratios, low heat release results in insufficient energy for reforming. On the other hand, if the O_2/C ratio is too high, there is the tendency for the emitted H_2 and CO to be oxidized into H_2O and CO_2, respectively. It is noted that increasing O_2/C ratio implies increasing heat release of oxidation, which in turn increases reforming temperature.

2.1. Measurement error analysis

The total measurement error (δ_k) includes a bias error (β) and a precision error (ε_k) such that [16]
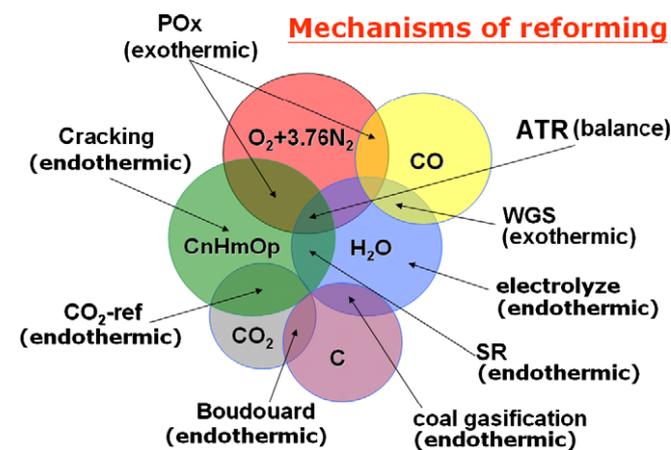

Fig. 2. Mechanisms of reforming.

Table 2
Operating parameters for methane and propane reforming.

	O_2/C_n	S/C_n	Flow rate (mol/min)
Methane	0.5–1	0–2	0.0404–0.4036
Propane	1.5–3	0–2	0.0202–0.2018
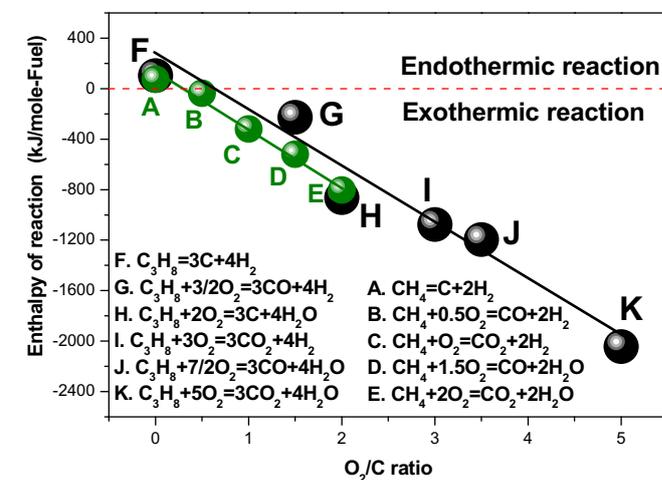
Fig. 3. Enthalpy of reaction of methane and propane reforming under different O_2/C ratios.

F. $C_3H_8=3C+4H_2$
G. $C_3H_8+3/2O_2=3CO+4H_2$
H. $C_3H_8+2O_2=3C+4H_2O$
I. $C_3H_8+3O_2=3CO+4H_2$
J. $C_3H_8+7/2O_2=3CO+4H_2O$
K. $C_3H_8+5O_2=3CO_2+4H_2O$

A. $CH_4=C+2H_2$
B. $CH_4+0.5O_2=CO+2H_2$
C. $CH_4+O_2=CO_2+2H_2$
D. $CH_4+1.5O_2=CO+2H_2O$
E. $CH_4+2O_2=CO_2+2H_2O$

Table 3
Measurement uncertainties.

H_2: ±1.01%
CO: ±0.51%
CO_2: ±0.51%
CH_4: ±0.50%
Temperature: ±2.6 °C
Air flow rate: ±2.31%
Fuel flow rate: ±2.22%

$$\delta_k = \beta + \varepsilon_k$$

The precision error is determined by taking N repeated measurements. The precision index of the average of a set of measurements is always less than that of an individual measurement according to

$$S_{\bar{x}} = \frac{S}{\sqrt{N}}$$

where S is the standard deviation of the N repeated measurements. The bias error is the systematic error considered to remain constant during a given test. The measurement uncertainty (U) with 95% confidence level can be given by the followed model:

$$U_{RSS} = [B^2 + (tS_{\bar{x}})^2]^{\frac{1}{2}}$$

where U_{RSS} is the measurement uncertainty derived by the root sum square (RSS) method, B is the bias limit, and t is set equal to two for large samples ($N > 30$).

According to the above analysis, the measurement uncertainties were estimated and shown in Table 3.

3. Results and discussion

The reforming processes encapsulated the endothermic and exothermic reactions. The control parameters included O_2/C ratio, fuel supply rate and the space velocity, all of which has an effect on temperature and gas residence time, the important factors of reforming. This section investigates the effect of reformate temperature on the ratio $H_2/(CO + CO_2)$, fuel conversion efficiency, the yield of hydrogen and carbon monoxide. The overall reforming thermal efficiency is also examined. Fig. 4 compares the reformate temperature on the ratio of $H_2/(CO + CO_2)$ under different fuel supply rates for methane and propane. In this figure, a line is drawn through six points with O_2/C ratios between 0.5 and 1.0 in 0.1 increments for each fuel flow rate. It is clear in the figure that
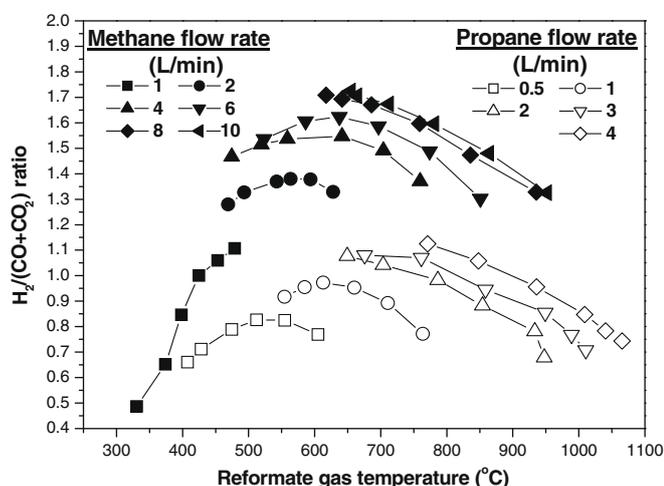
the combination of high O_2/C ratio and high reformate temperature, with low methane supply rate results in a good $H_2/(CO + CO_2)$ ratio. The combination of high fuel supply rate and low O_2/C ratio also attains good $H_2/(CO + CO_2)$ ratio. However, for the low methane rate of 1 L/min, the $H_2/(CO + CO_2)$ ratio is obviously lower than other flow rates despite increasing with increasing O_2/C value. The reason is that the reformate temperature is low due to less fuel oxidization and therefore less oxidation heat, which causes poor conversion of fuel and different selectivity of reformate gas. It is clear from the experimental results, the yield of H_2 and CO is low; but relatively high yield of CO_2 is obtained for the conditions of low methane supply rate and low O_2/C ratio, i.e., low reformate temperature. As the temperature increases, the CO_2 yield increase is limited, but the H_2 yield continues to increase. Hence, the ratio of $H_2/(CO + CO_2)$ is low as the reformate temperature is relatively low. However, as the temperature increases, the increase in $H_2/(CO + CO_2)$ ratio is more apparent before peaking. It is also evident from the figure that methane gives an overall higher $H_2/(CO + CO_2)$ ratio than propane, and that its maximum hydrogen production is almost twice. The main reason is the higher H/C ratio of methane, the value of 4, the highest of all hydrocarbons, compared to 2.66 for propane. In general, the peak $H_2/(CO + CO_2)$ ratio for both types of fuel occurs between 600 °C and 800 °C.

Fig. 5 compares the effect of reformate temperature on fuel conversion efficiency with methane and propane under different supply rates. It is evident that good conversion efficiency is achieved with propane even for low supply rates. The main reason is that the release heat produced by per unit molar of propane is higher and therefore under low supply rate, all tested O_2/C ratios could generate high oxidation heat for reforming. It is also apparent from the figure that the reformate temperature for complete conversion of methane is lower at 850 °C compared to 1065 °C for propane.

Fig. 6 shows the relationship between reformate temperature and the yield of H_2 and CO as well as thermal efficiency using methane and propane under different supply rates. It is clear from the figure that the maximum yield when using methane is obtained at a temperature between 700 °C and 800 °C, whilst the maximum yield when using propane is higher at between 800–900 °C. After a peak, the hydrogen production appears to decrease with increasing reformate temperature, the reason of which can be related to the conversion efficiency shown in Fig. 5. After all the supplied fuel is combusted at high reformate temperature, excess oxygen from the high O_2/C ratio will then react with the produced hydrogen and carbon monoxide to form water and carbon dioxide,

Fig. 4. Relationship of reformate gas temperature and $H_2/(CO + CO_2)$ under different intake flow rates of methane and propane.
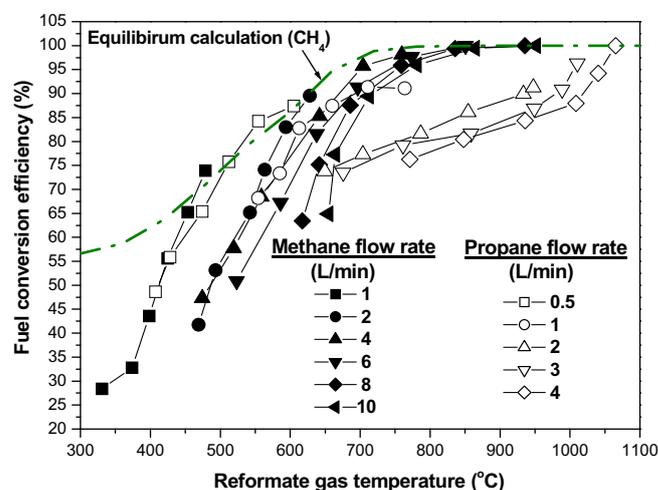
Fig. 5. Relationship between reformate gas temperature and fuel conversion efficiency under different flow rates for methane and propane.
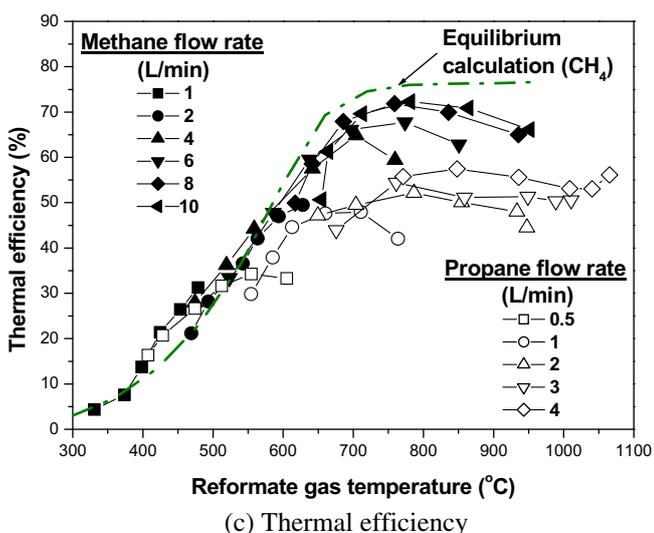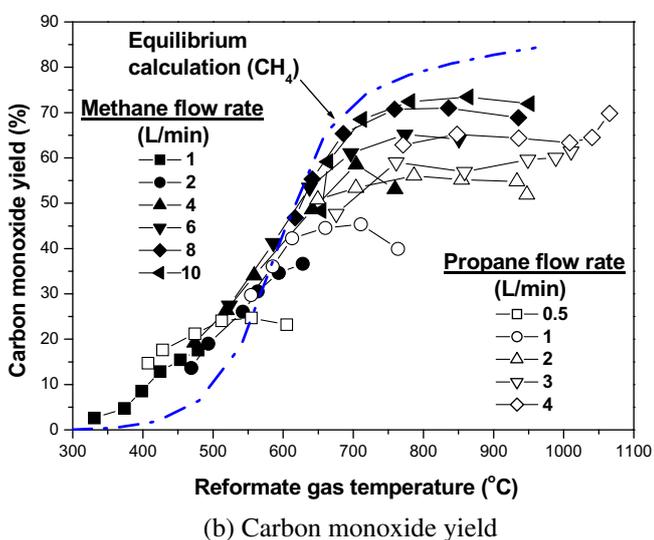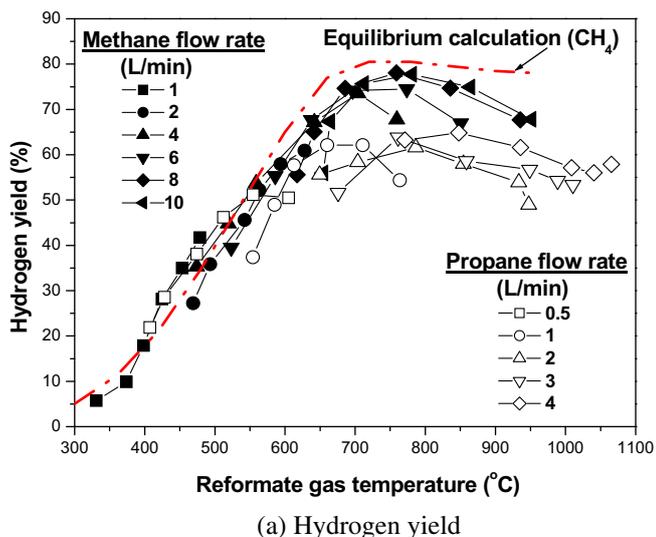
(a) Hydrogen yield

(b) Carbon monoxide yield

(c) Thermal efficiency

Fig. 6. Relationship between reformate gas temperature and H_2, CO yield and thermal efficiency under different gas flow rates.

Table 4
Reforming characteristics of methane and propane.

Fuel type	CH_4		C_3H_8	
Input (mol/min)	0.4036		0.2018	
O_2/C	0.8		0.5	
S/C	n/a	0.5	n/a	0.5
Reactions	POX	OSR	POX	OSR
Reaction temp. (°C)	743	785	794	739
H_2/CO ratio	2.14	3.02	1.32	2.12
H_2 + CO (vol %)	41.44	39.93	42.63	40.15
Conversion (%)	95.99	95.59	78.60	79.50
H_2 selectivity (%)	81.00	89.88	83.00	93.61
CO selectivity (%)	75.00	59.42	83.50	58.75
H_2 yield (%)	77.75	85.92	65.00	74.45
CO yield (%)	72.44	56.80	65.00	46.73
Syngas flow rate (L/min)	22.57	22.82	22.75	22.12
H_2 + CO output power, LHV (kW)	3.90	3.86	4.04	3.79
Thermal efficiency (%)	72.01	71.27	58.50	54.88
H_2 output power, LHV (kW)	2.52	2.77	2.13	2.43
Enthalpy of reaction (kJ/mol)	−194.7	−220.4	−448.0	−483.1
Energy loss ratio (%)	24.34	25.06	21.93	23.62

to a certain stage where it graduals off. The trend of the thermal efficiency is similar to that of the yield of hydrogen and carbon monoxide. Thermal efficiency is revealed to be the best at a temperature of approximately 750 °C for methane whilst it is 850 °C for propane. Between the two, using methane as fuel results in better thermal efficiency as well as lower reforming temperature, preventing high sustained operating temperature at the catalyst that may damage its activity. The experimental results were compared with theoretical equilibrium calculations using the commercialized software HSC for a methane supply rate of 10 L/min and an O_2/C ratio of 0.8. In terms of the yield of H_2 and CO and thermal efficiency, a favorable trend has been achieved.

From the above tests, the reforming characteristics of methane and propane have been demonstrated. This section further investigates the effect of these characteristics. The reforming characteristics of methane and propane under optimum operating conditions are summarised in Table 4 for comparison. The fuel input rate of methane is 0.4036 mol/min and the O_2/C ratio is 0.8. These are 0.2018 mol/min and 0.5, respectively, for propane. The fuel input rate is selected to produce approximately the same theoretical hydrogen output rate for both fuels. From Table 4, it is clear that the conversion efficiency of methane is far superior to propane. This results in higher yield of H_2 and CO due to higher selectivity of H_2 and CO for methane. The maximum hydrogen yield is as high

respectively. It is also evident in the figure that the yield of carbon monoxide rapidly increases with increasing reformate temperature
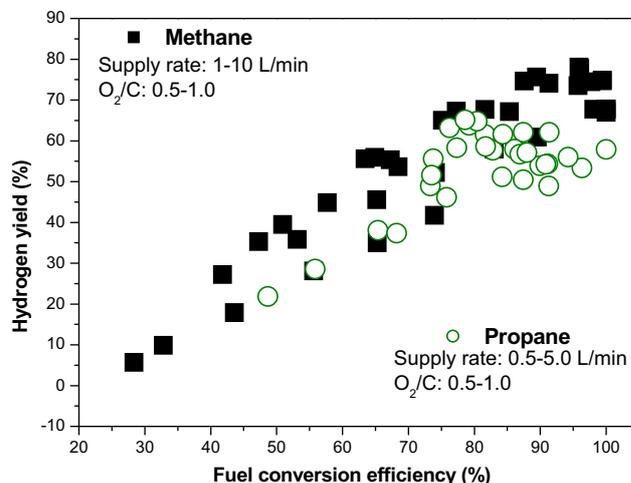

Fig. 7. Relationship between fuel conversion efficiency and hydrogen yield.
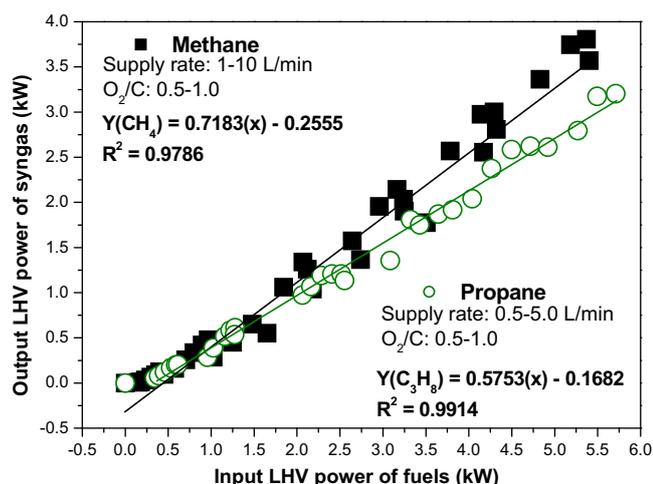
Fig. 8. Relationship between input power of fuels and output power of syngas (LHV).

as 77.75%, equivalent to a hydrogen output rate of 15.40 L/min, compared to 13.04 L/min when using propane.

Fig. 7 shows the effect of fuel conversion efficiency on hydrogen yield. Overall, at a given conversion efficiency, methane results in better hydrogen yield. As the conversion efficiency increases, the hydrogen yield with methane increases nearly proportionally whilst with propane, the yield increases up to about 85% fuel conversion efficiency where it then gradually reduces. Fig. 8 compares the reforming performance of these two fuels. The x-axis represents the equivalent power of the fuel supply rate that is directed into the reaction chamber whilst the y-axis is the output power converted from the flow rate of H_2 and CO. It is apparent that under a given fuel intake rate, methane results in a better thermal efficiency than propane. This becomes more pronounced with increasing fuel supply rate.

The reforming performance of methane and propane using the partial oxidation reforming process with steam addition was further compared in Table 4. It is clear that with a setting of O_2/C ratio of 0.8 and S/C ratio of 0.5, using methane in oxidative steam reforming instead of partial oxidation, the hydrogen yield increases from 77.75% to 85.92%. In comparison, with an O_2/C ratio of 0.5 and an S/C ratio of 0.5, using propane gives a higher hydrogen yield of 74.45% compared to 65.00% obtained through partial oxidation. Despite the more efficient hydrogen yield, the corresponding thermal efficiency is slightly lower for oxidative steam reforming. The possibility is due to the poor gain in H_2 by CO consumption through the water gas shifting reaction. In other words, the CO conversion is pronounced but the H_2 yield is only improved marginally, therefore resulting in a relatively lower overall thermal efficiency.

4. Conclusions

The reforming performance of two low carbon fuels, namely, methane and propane, has been compared in this paper. Experiments have shown that by combining a plasma converter with a catalyst, a good yield of hydrogen-rich gas was produced. The re-

sults obtained in the tests could be used as the future application of reforming processes with other types of fuels, such as natural gas and LPG. The experiment results have demonstrated temperature was an important factor in the overall reforming performance. It has also been revealed that the $H_2/(CO + CO_2)$ ratio obtained with methane and propane, increased with increasing temperature, with the best ratio for methane occurring at 650 °C. Fuel conversion efficiency was also found to increase with increasing reformate temperature. When using methane, the best thermal efficiency was 72.01% at around 750 °C. The corresponding values for propane were 58.50% at around 800 °C. Hydrogen yield was shown to improve under an appropriate steam addition at S/C = 0.5, giving a hydrogen yield of 85.92% compared to 77.75% without steam addition, both with the optimum setting for reforming methane.

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