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Reforming options for hydrogen production from fossil fuels for PEM fuel cells

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Abstract

PEM fuel cell systems are considered as a sustainable option for the future transport sector in the future. There is great interest in converting current hydrocarbon based transportation fuels into hydrogen rich gases acceptable by PEM fuel cells on-board of vehicles. In this paper, we compare the results of our simulation studies for 100 kW PEM fuel cell systems utilizing three different major reforming technologies, namely steam reforming (SREF), partial oxidation (POX) and autothermal reforming (ATR). Natural gas, gasoline and diesel are the selected hydrocarbon fuels. It is desired to investigate the effect of the selected fuel reforming options on the overall fuel cell system efficiency, which depends on the fuel processing, PEM fuel cell and auxiliary system efficiencies. The Aspen-HYSYS 3.1 code has been used for simulation purposes. Process parameters of fuel preparation steps have been determined considering the limitations set by the catalysts and hydrocarbons involved. Results indicate that fuel properties, fuel processing system and its operation parameters, and PEM fuel cell characteristics all affect the overall system efficiencies. Steam reforming appears as the most efficient fuel preparation option for all investigated fuels. Natural gas with steam reforming shows the highest fuel cell system efficiency. Good heat integration within the fuel cell system is absolutely necessary to achieve acceptable overall system efficiencies.
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1. Introduction

Proton exchange membrane fuel cell (PEMFC) powered vehicles offer the potential for high efficiency and reduced emissions. Primary PEMFC transport applications include light duty (50–100 kW) and medium duty (200 kW) vehicles. PEMFCs require a high purity hydrogen source for operation. Hence, the projected commercialization of PEMFC powered vehicles requires a readily available hydrogen source, which is either used directly or is produced in an on-board fuel processor [1,2]. Hydrogen can be produced by reforming a hydrocarbon fuel into a hydrogen rich gas mixture. Hence, fuel processing represents a significant challenge to the commercialization of PEMFCs. Natural gas, gasoline and diesel

appear as attractive hydrocarbon sources for fuel processors due to their existing distribution and supply infrastructure to generate hydrogen [3–13].

It is increasingly recognized that the fuel processing subsystem can have a major impact on overall fuel cell system efficiency and costs, and wide-spread implementation [14]. Reforming is being intensively developed for both on-board and off-board applications.

Partial oxidation (POX), autothermal reforming (ATR) and steam reforming (SREF) are the primary methods used in reforming hydrocarbons to produce hydrogen for use in PEM fuel cells.

Partial oxidation [17] and autothermal reforming [18,19] processes do not require indirect heating in contrast to steam reforming. Moreover, they offer faster startup time and better transient response. However, the product quality is poor due to low hydrogen concentrations, 70–80% for steam reform-

Nomenclature

AC	air compressor
ATR	autothermal reformer
C	chimney
COM	combustor
E	expander
E_i	energy of unit i
FC	fuel cell
FS	fuel splitter
FV	fuel vaporizer
HTS	high temperature shift reactor
HTS-C	HTS cooler
LHV	lower heating value (MJ kmol^{-1})
LTS	low temperature shift reactor
LTS-C	LTS cooler
m_i	mass flow rate (kg h^{-1})
P	power (W or kW)
PC	power (compressor) (kW)
PE	power (expander) (kW)
PEMFC	polymer electrolyte membrane FC
PEM-C	PEM FC cooler
PEMAC	AC power (PEM fuel cell) (kW)
POX	partial oxidation reactor
PROX	preferential oxidation reactor
PROX-C	PROX cooler
P1	fuel pump
P2	water pump
P3	cooling water pump
SG	steam generator
SREF	steam reformer
SREF-HE	SREF heat exchanger
T	splitter or temperature (°C)
V	voltage (V)
WFM	water fuel mixer
WT	water tank
1	SREF inlet
2	SREF exit
3	HTS exit
4	LTS exit
5	PROX exit
6	FC exit
η	efficiency

POX, SREF and ATR systems using gasoline as a feed and concluded that ATR was optimum from an efficiency standpoint. Doss et al. [27] studied process variables to optimize the efficiency of an ATR-based fuel processor integrated into an overall fuel cell system.

The reformed fuel often contains other gases such as carbon monoxide (CO) that are detrimental to PEMFC operation. The CO contained in the reformate must be further reduced to ca. 10 ppm prior to feeding to the PEM fuel cell. Various options for CO reduction include water–gas shift, preferential oxidation of CO, methanation, membrane separators and combinations of these with the reforming reactor [28].

In practice, the PEMFC system is usually fairly complex. Key components typically include a fuel processor, a fuel cell stack (which is typically made up of a large number of individual cells) and a power conditioner for converting the DC output of the fuel cell stack to AC power at the required voltage and frequency. Auxiliary systems such as pumps, compressors, expanders and blowers are required for fuel, air and water management. Heat exchangers are used for heat integration within the PEMFC system [29].

Steam reforming (SREF), partial oxidation (POX) and autothermal reforming (ATR) are the three major hydrocarbon-reforming technologies for PEM fuel cells. The primary purpose of this paper is to identify favorable operating conditions at which the selected fuels are converted to hydrogen rich gas mixtures via SREF, POX and ATR processes at reasonable fuel reforming efficiencies. Selected reforming options are integrated with FCs, and the resulting PEMFC systems are analyzed. Heat integration within the PEMFC systems is of utmost importance since the consumption of thermal energy is a key issue in the design of the reforming systems. Balance of plant equipment has also to be included in the PEMFC system analyses. The results provide a comparative evaluation basis for the selected fuel/fuel reforming options along with a starting point for experimental research.

2. Methodology

Natural gas, gasoline and diesel type hydrocarbon mixtures have been studied as three different sources for hydrogen production. The chemical compositions of the liquid hydrocarbon fuels used in the simulation studies are provided elsewhere [31–33]. Table 1 summarizes the chemical components of the natural gas fuel. The average molecular weights are around 16.4 kg kmol^{-1}, 94.5 kg kmol^{-1} and 215.3 kg kmol^{-1} for the natural gas, gasoline and diesel fuels, respectively. All simulations of this study are based on these compositions.

The use of chemical flow-sheeting software has become an integral part of the evaluation of the performance of fuel cell systems [16,30]. The steady state simulation computer code of the Aspen-HYSYS process-modeling tool has been

ing versus 40–50% for partial oxidation and autothermal reforming on a dry basis. Compared with partial oxidation and autothermal reforming, catalytic steam reforming offers higher hydrogen concentrations. The steam reforming reaction, on the other hand, is a highly endothermic reaction and requires heating [11]. Steam reforming of hydrocarbons for fuel cell applications have been discussed by Gunardson [20], Rostrup-Nielsen [21] and Armor [22] for stationary H$_2$ plants in the gas industry, and by Clarke et al. [23], Dicks [24], and Privette [25]. Docter and Lamm [26] compared

Table 1
Chemical composition of natural gas

Component	Mass fractions (wt%)	Molar fractions (mol%)
Methane	95.2	97.4
Nitrogen	4.1	2.4
DiBZThiphene	0.1	0.0
Ethane	0.2	0.1
i-Butane	0.0	0.0
n-Butane	0.1	0.0
Propane	0.2	0.1
Total	100	100

Table 2
The investigated ranges of reformer operating conditions

	Temperature (°C)	Pressure (bar)	S/C	O/C
ATR	600–900	3	2.0–3.5	0.25–2.0
PRE-SREF	500–550	3	2.0–3.5	–
SREF	700–850	3	2.0–3.5	–
POX	800–1100	3	–	0.25–2.0

Table 3
The PEM fuel cell characteristics (e⁻: electron)

Anode reaction	$H_2 \rightarrow 2H^+ + 2e^-$
Cathode reaction	$\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O$
Fuel utilization (%)	80
Fuel cell outlet temperature (°C)	70
Pressure (bar)	3
Average cell voltage (mV)	750
Active area (m²)	0.04
Stack cooling media	Water
Air utilization (%)	50
Current density (mA cm⁻²)	249
DC output power (kW)	100
Number of single cells	1250

utilized along with conventional calculations for fuel cell systems.

The simulated PEM fuel cell system consists of the following sections:

- fuel processing section;
- PEM fuel cell section;
- auxiliary units.

Fuel processing consists of reforming and clean-up sections (Fig. 1). The reforming section contains the reforming reactor(s): an autothermal (ATR) or two steam reforming units (PRE-SREF and SREF), or a partial oxidation (POX) reactor. The clean-up section is made up by high and low temperature shift reactors (HTS and LTS) and the preferential oxidation reactor (PROX).

For all cases, all reactors are simulated to operate under equilibrium conditions. The thermodynamic equilibrium system calculations are based on minimizing the Gibbs free energy. All reactor simulation calculations have been performed keeping "$T_{reactor}$" almost constant taking heats of reaction into account.

The pressure is kept constant at 3 bar. The S/C, O/C ratios and operation temperatures of reactors are changed parametrically to determine the best operation parameters. The limitations set by the catalysts and hydrocarbons involved are also considered. The simulation code is capable to calculate the steady state product compositions taking into account the incoming stream compositions under the defined operation conditions. Table 2 presents the ranges of operating parameters investigated in the simulation studies.

The aim is to convert as much as the hydrogen in the fuel into hydrogen gas at acceptable yields in an efficient manner while decreasing CO and CH₄ formation. Lower S/C ratios favor soot and coke formation, which is not desired in catalytic steam and autothermal reforming processes. A

considerably wide S/C ratio (2.0–3.5) range has been selected to see its effect on hydrogen yield and CO formation [9–38]. A similar approach has been adopted for O/C ratio which is changed between 0.5 and 2.0.

The PEM fuel cell section consists of the following components:

- fuel cell stack;
- DC/AC converter.

The PEM fuel cell module has been simulated using the PEM fuel cell characteristics presented in Table 3. All the characteristic figures are calculated for a fuel cell stack with 1250 cells and a cell active area of 400 cm². Fig. 2 shows the polarization curve based on the data from the real fuel cell unit [39].

Auxiliary units comprise pumps, compressor, expander, heat exchangers, heaters coolers and combustor. Their properties summarized in Table 4 are based on commercially available units. The heat exchanger model chosen for our analysis is the ideal (Ft = 1) counter current weighted design model. The log mean temperature difference (LMTD) correction factor, Ft, is calculated as a function of the number of shell passes and the temperature approaches.

Extensive heat integration is sought within the study to achieve acceptable overall system efficiency levels. The over-
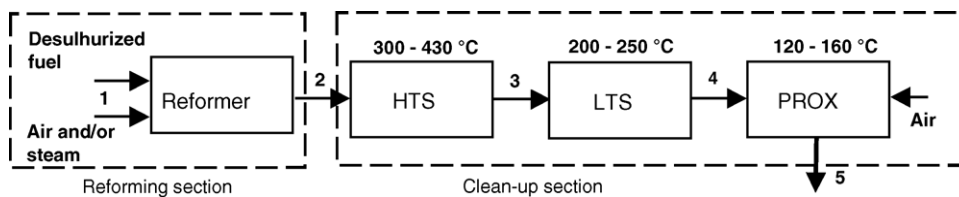

Fig. 1. Schematic diagram of the fuel reforming and clean-up sections.
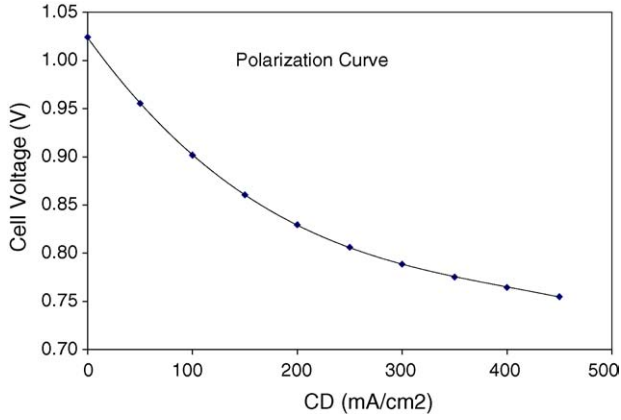
Fig. 2. Polarization curve.

Table 4
Auxiliary system component data

Component	Parameter	Value
Fuel pump	Adiabatic efficiency (%)	65
Water pump	Adiabatic efficiency (%)	75
Cooling water pump	Adiabatic efficiency (%)	75
Compressor	Adiabatic efficiency (%)	70
Expander	Adiabatic efficiency (%)	75
Heat exchangers	Minimum temperature approach (°C)	25
Combustor	Outlet temperature (°C)	650
Chimney	Outlet temperature (°C)	155
DC/AC converter	Conversion efficiency (%)	98

all system efficiency ($\eta_{net.el}$) is calculated as the product of fuel processing (η_{FP}), PEM fuel cell (η_{FC}) and auxiliary ($\eta_{Aux.}$) system efficiencies.

$$\eta_{net.el} = \eta_{FP}\eta_{FC}\eta_{Aux.}$$

The fuel processing efficiency covers the section from the hydrocarbon feed section to the fuel cell including all reforming and clean-up reactors and auxiliary equipment; Table 5 summarizes the major calculation steps. The mass flow rates, temperature and pressure values of the mass streams utilized in efficiency calculations are presented in Table 6 while Fig. 3 shows their position in the fuel cell system. Fig. 3 is a simplified sketch of the actual PEMFC system studied.

Table 5
The formulae of the efficiency calculations for various components and subsystem

m_i = mass flow rate (kg h^{-1})
$\eta_1 = (m_{103}LHV_{103})/(m_F LHV_F)$
$\eta_{PRE-SREF} = (m_{401}LHV_{401})/(m_{400}LHV_{400})$
$\eta_{SREF} = (m_{404}LHV_{404})/(m_{402}LHV_{402})$
$\eta_{HTS} = (m_{406}LHV_{406})/(m_{404}LHV_{404})$
$\eta_{LTS} = (m_{408}LHV_{408})/(m_{406}LHV_{406})$
$\eta_{PROX} = (m_{410}LHV_{410})/(m_{408}LHV_{408})$
$\eta_2 = \eta_1\eta_{PRE-SREF}$
$\eta_3 = \eta_2\eta_{SREF}$
$\eta_4 = \eta_3\eta_{HTS}$
$\eta_5 = \eta_4\eta_{LTS}$
$\eta_6 = \eta_5\eta_{PROX} = \eta_{FP}$

The PEM fuel cell (η_{FC}) efficiency depends on hydrogen utilization ratio (U_{H_2}) and stack voltage and DC/AC conversion efficiencies. Table 7 presents the basics of efficiency correlations used in the PEM fuel cell efficiency (η_{FC}) calculations

$$\eta_{FC} = \eta_{stack\ voltage}\eta_{DC/AC}$$

The auxiliary system efficiency ($\eta_{Aux.}$), is calculated as follows:

$$\eta_{Aux.} = 1 + \frac{(P_E - P_{parasitic})}{P_{PEMAC}}$$

$$P_{parasitic} = \frac{P_{P1} + P_{P2} + P_{P3} + P_C}{\eta_{motor}}, \qquad \eta_{motor} = 0.90$$

3. Results and discussion

Fuel processing and net electrical efficiencies of natural gas and gasoline/diesel fuels for the investigated fuel-reforming options are presented in Table 8. The simulation results indicate that the fuel processing efficiencies decrease in the order of steam reforming > autothermal reforming > partial oxidation for both gasoline and diesel fuels. Steam reforming appears as the most promising fuel reforming option based on fuel processing efficiencies. Only minor differences have been observed in terms of efficiencies of the selected gasoline and diesel fuels [40,41].

Natural gas shows higher fuel processing efficiencies then the liquid fuels, hence, also higher overall system efficiencies. The highest fuel processing efficiency is achieved for the steam reforming of natural gas, namely 98%. The same option gives a maximum net electrical efficiency at 48% (Table 9). Hence, the natural gas with steam reforming is about 14% more efficient than its liquid fuel counterparts based on steam reforming (Table 8).

Here, we present the results of the most efficient option, namely natural gas with steam reforming. The major units of the Aspen-HYSYS simulation for natural gas steam reforming based fuel cell system are presented in Fig. 3. In contrast to liquid fuel systems natural gas based systems do not require the pre-reformer unit due to their high lower molecular weight hydrocarbon, namely CH_4 content [42–44].

The operation parameters of the SREF are of utmost importance to achieve the desired high hydrogen and low CO content product gases along with acceptable fuel conversion efficiency levels. The steam to carbon ratio range studied is between 2 and 4 while the operating temperature has been changed between 500 °C and 850 °C (Fig. 4). The effect of S/C ratio becomes more and more pronounced as the operating temperature increases. The S/C ratio at 3.5 appears to achieve the conversion requirements for temperatures around 800 °C. A decrease of the S/C ratio decreases both the efficiency as well as the hydrogen content of the product gases. The concentration of the unconverted CH_4 in

Table 6
Simulation results for selected system points calculated under the prescribed operating conditions applied in this study

| Stream	Fuel	Water	Air	400	301	302	402	403	404
Temperature (°C)	25.0	331.1	192.3	25.0	192.3	192.3	520.0	800.0	350.0
Pressure (bar)	500.0	406.2	325.0	100.0	325.0	300.0	388.7	388.7	360.0
Molar flow (kmol h^{-1})	0.9	2.6	0.1	11.3	0.1	11.3	3.4	4.9	4.9
Mass flow (kg h^{-1})	14.6	47.6	1.9	327.4	1.9	325.5	60.0	60.0	60.0
	405	406	407	408	409	410	500	501	600
Temperature (°C)	422.4	200.0	230.4	120.0	151.9	69.5	70.0	70.0	650.0
Pressure (bar)	350.0	342.5	332.5	325.0	315.0	300.0	300.0	300.0	257.2
Molar flow (kmol h^{-1})	4.9	4.9	4.9	4.9	4.9	4.2	1.8	13.4	11.6
Mass flow (kg h^{-1})	60.0	60.0	60.0	60.0	61.9	48.1	43.4	348.0	329.8
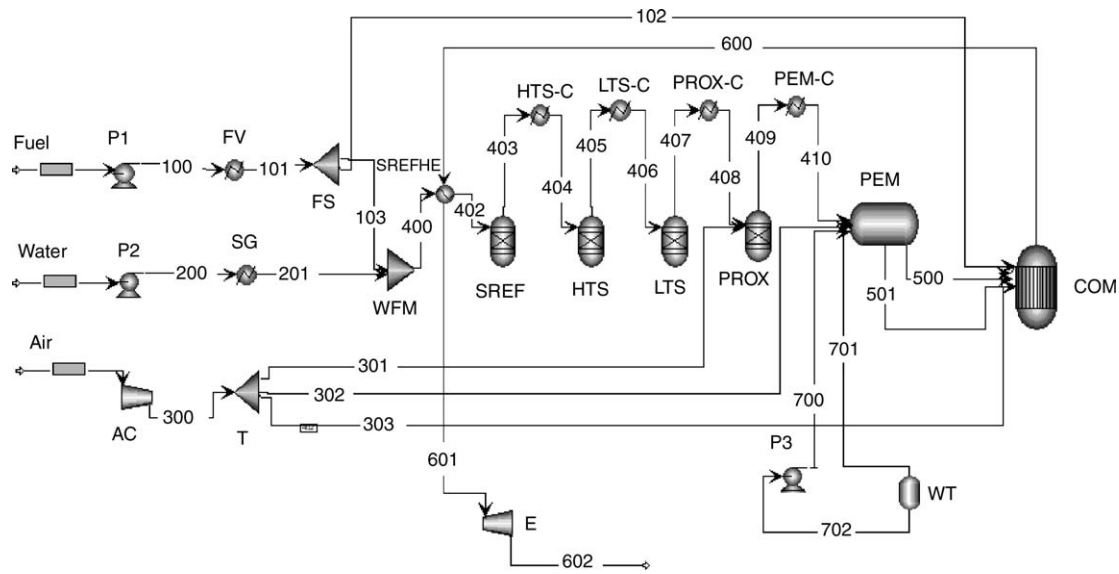
Fig. 3. Major fuel cell system units of the Aspen-HYSYS simulation for natural gas with steam reforming.

the steam reformer products steadily decreases as the temperature increases from 500 °C to 800 °C. Therefore, the operation parameters of the SREF are selected as 800 °C and S/C = 3.5. The product compositions for all the other fuel processing reactors following the steam reformer are presented in Figs. 5 and 6.

The SREF based fuel-processing, fuel cell, auxiliary and overall system efficiencies of the investigated fuels are pre-

sented in Table 9. The values indicate that natural gas with steam reforming is the best option while there are only minor differences regarding the investigated liquid fuels. The slight differences observed between gasoline and diesel options are primarily due to the better heat integration achieved with the latter.
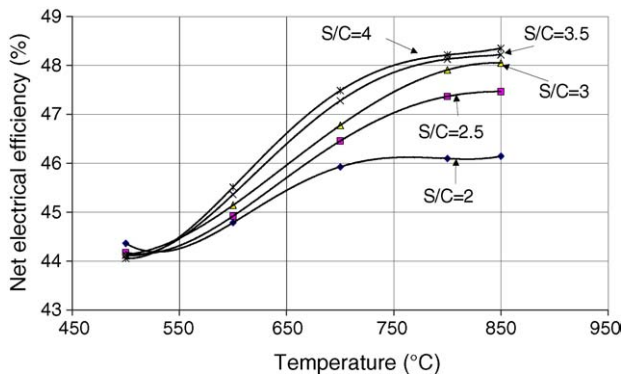

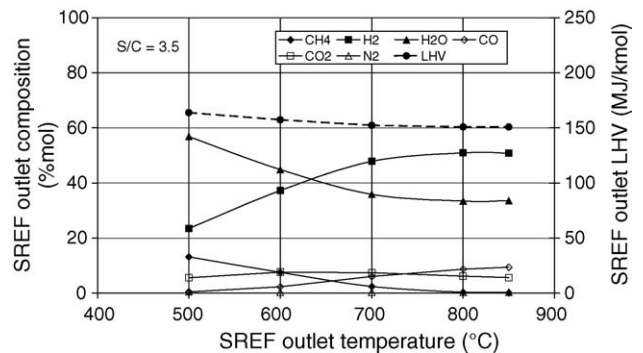Fig. 4. Effect of the S/C ratio on overall system efficiency.

Fig. 5. The molar compositions and LHV values of the steam reformer products.
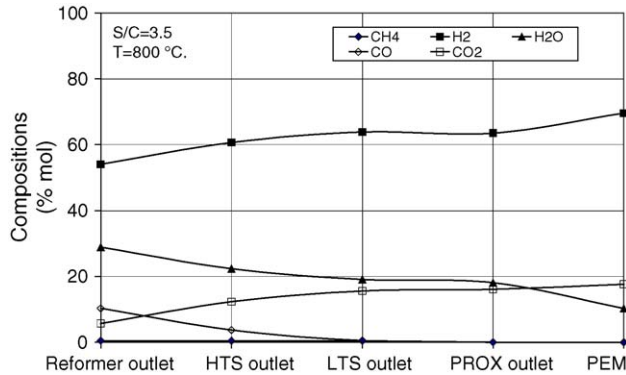
Fig. 6. Product compositions of fuel preparation reactors.

Table 7
The basic correlations for PEM fuel cell efficiency (η_{FC}) calculations

$\eta_{\text{stack voltage}} = V_{\text{cell}} U_{H_2}$
$V_{\text{cell}} = f(I_{\text{current}})$
$I_{\text{current}} = I_{\text{calculated}} / A_{\text{active}}$
$A_{\text{active}} = \text{cell active area (m}^2)$
$I_{\text{calculated}} = H_{2\text{ consumed}} \times 2 / (1.04 \times 10^{-8} n_{\text{cell}} \times 3600)$
$(\eta_{DC/AC}) = 0.98$
$U_{H_2} = 0.80$

Table 8
Overall fuel processor and net electric efficiency for natural gas

Fuel	Process	Efficiency (η)	With heat integration	Without heat integration
NG	SREF (S/C = 3.5)	η_{FP}	98	89
		$\eta_{net.el}$	48	39
Gasoline/ diesel	SREF (S/C = 3.5)	η_{FP}	86	–
		$\eta_{net.el}$	42	–
	ATR (S/C = 2.5, O/C = 0.5)	η_{FP}	86	–
		$\eta_{net.el}$	37	–
	POX	η_{FP}	74	–
		$\eta_{net.el}$	31	–

Table 9
SREF based fuel processing, fuel cell, auxiliary and overall system efficiencies

Fuel	η_{FP}	η_{FC}	$\eta_{Aux.}$	$\eta_{net.el}$
Natural gas	98.1	50.6	97.0	48.2
Gasoline	85.5	50.6	95.1	41.1
Diesel	86.3	50.6	97.1	42.4

4. Concluding remarks

Natural gas appears as the best fuel for hydrogen rich gas production due to its favorable composition from lower molecular weight compounds. Steam reforming and autothermal reforming appear as the most competitive fuel processing options (Table 8) in terms of fuel processing efficiencies. POX

shows the lowest fuel processing efficiency level. Among the options studied the highest fuel processing efficiency is achieved with natural gas steam reforming at about 98%.

High PEMFC system efficiency levels can be achieved only with intensive heat integration within the PEMFC systems (Table 8). Hence, heat integration system studies are of utmost importance along with the development of novel reforming catalysts, clean-up systems and PEMFC components if on-board hydrogen production is desired.

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