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## Plasma fuel reforming: a critical review

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### Abstract

In this work, a recent development of plasma fuel reforming was critically reviewed. The fundamental characteristics, the reforming efficiency and the energy efficiency of the nonthermal plasma reforming of hydrocarbon gases and liquid fuels, and the thermal plasma reforming of liquid and solid fuels will be discussed in detail.

**Keywords:** Plasma reforming; Natural gas; Coal; Solid waste; Liquid fuel

## 1 Introduction

Recent development of low pollution, emission and high thermal efficiency fuel requirements generated a new application area called the plasma clean fuel reforming. In this work, the fundamental reforming characteristics and the energy efficiency of the reforming by the thermal and non-thermal plasmas for solid, liquid and gaseous fuels are critically reviewed. The focus of the fuels considered is:

- Gaseous Fuel: Natural Gas (Methane, Propane), Waste Gases, etc.
- Liquid Fuel: Gasoline, Diesel, Heavy Oil, Waste Oils, Bio-Oils etc.
- Solid Fuel: Coal, Plastic and Rubber Wastes, Tar Sand, Sludges, Methane Hydride, Biomass, etc.

The plasma systems considered are:

- A. Thermal Plasma: DC, RF and Microwave Plasma Torches; DC, AC single-phase and three-phase arcs, etc.

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- B. Non-Thermal Plasma: Electron Beam, Flow Stabilized DC Coronas, Pulsed Corona, AC Barrier Discharge-Packed Bed, Surface and Silent Discharges, etc.
- C. Plasma-Catalyst: Hybrid and Superimposed Systems.

## 2 Gaseous fuel reforming

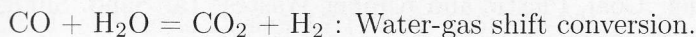
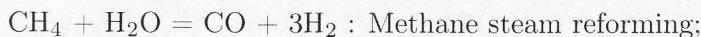
For a co-generation system using a fuel cell, the hydrogen fuel is normally generated from hydrocarbon gases such as butane, propane, methane, or natural gas since they are already used in towns as gases for home heating or coking. Using proton exchange membrane (PEM) type fuel cells up to 80% efficiency can be achieved if CO and soot free hydrogen dominant syngases can be produced. CO is the main poison for PEM type fuel cells [1]. At the moment, shift conversion of natural gas reformer has been used. However, this method requires high pressures and high temperatures.

### 2.1 Conventional and plasma reforming of natural gas

Typical natural gas consists of approximately 94 to 95% CH<sub>4</sub> (methane), 2 to 3% C<sub>2</sub>H<sub>6</sub> (ethane), 0.1 to 0.3% C<sub>3</sub>H<sub>8</sub> (propane), 1.5 to 2% N<sub>2</sub>, 0.5 to 0.6% CO<sub>2</sub> with traces of iso- and n-pentane (0.01 – 0.02% C<sub>5</sub>H<sub>12</sub>), and C<sub>6</sub>H<sub>14</sub> and sulphur (S) (7 to 8 mg/m<sup>3</sup> in North America). Some European natural gas may contain a small percentage of H<sub>2</sub>S. Its relative density is on average 0.582 that of air, and its heating value is 38 MJ/m<sup>3</sup> (or 1020 BTU/CF) [43]. Hence, any oxidation processes at elevated temperatures generate thermal and fuel NO<sub>x</sub> as well as SO<sub>2</sub> and CO as unwanted by-products. Thus, most processes start with the removal of all other natural gas components, then convert methane using one of the following methods:

#### A. Steam reforming

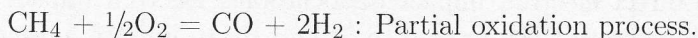
Methane is reformed by steam as follows:



Hence, a mixture of CO, CO<sub>2</sub> and H<sub>2</sub> can be generated at high gas temperature conditions without a catalyst or producing methanol mixtures with a catalyst [2-4].

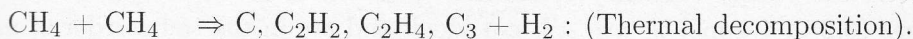
#### B. Partial oxidation process

Methane can be reformed under high gas temperature conditions with or without a catalyst [5, 6], as follows:



### C. Thermal decomposition

Based on thermodynamic calculations, methane can thermally dissociate at approximately  $2$  to  $6 \cdot 10^3$  K as follows:



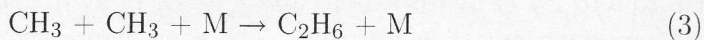
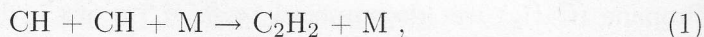
Based on the Senkin code simulation, a gas temperature of approximately  $3 \cdot 10^3$  K is the optimum condition for Hydrogen production [7, 8].

### D. Arc plasma process

Arc plasma, or thermal plasma, can generate extremely high gas temperatures (up to  $2 \cdot 10^4$  K), which normally cannot be generated by other combustion or electrical heating processes. Hence, an ac arc plasma reactor is commonly used for methane reforming with suitable quenching processes as follows [9-12]:



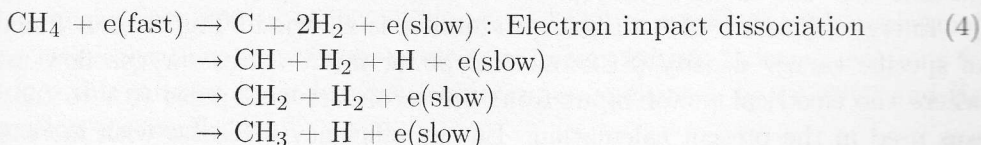
However, this process also generates:



and trace  $\text{C}_3\text{H}_6$  as by-products.

### E. Non-thermal plasma process

Unlike arc or thermal plasmas, non-thermal plasmas generated by corona discharges, microwave discharges or electron beams, i.e. below combustion temperature ( $\approx 2300$  K), initiate the majority of processes by an energetic electron (mean electron temperature  $T_e = 1$  to  $10^2$  eV) as follows [13-16]:



where the total reaction rate and branching fraction depend on the electron energy. Besides direct electron impact dissociations, electron impact dissociative ionization will generate  $\text{CH}_x^+$  ( $x = 0$  to 5) and  $\text{H}_2$  to initiate ion-molecule reactions to form  $\text{CH}_y$  ( $y = 0$  to 3) and  $\text{H}_2$ . Hence, a mixture of  $\text{H}_2$ ,  $\text{C}_2\text{H}_z$  ( $z = 2, 4, 6$ ) and  $\text{C}_3\text{H}_m$  ( $m = 3 - 6, 8$ ) can be generated [1-30]. With a mixture of  $\text{CH}_4\text{-O}_2$  or  $\text{CH}_4\text{-CO}_2$ , reforming gas with additional  $\text{CO}$  or  $\text{CH}_3\text{OH}$  can also be generated. However, the energy efficiency of methane conversion obtained by non-thermal plasmas (0.2 to 4 mmol/J) are still not ideal, and are in the same order of magnitude as the ac arc (2 to 5 mmol/J) or high temperature oxidation processes [1-30]. A non-thermal plasma process, based on a flow stabilized corona discharge system, is used for propane reforming. Propane was used instead of natural gas, since propane contains more hydrogen per molecule compared with methane, and is still in gas phase near atmospheric pressure. Moreover, hydrogen production from hydrocarbon, by non-thermal plasmas, increases with increasing  $C_x$  [31]. However, due to the high  $C_x$ , soot formation must be considered [32].

## 2.2 Plasma reforming of propane

Typical plasma reformed gas composition as a function of primary side power (charging voltage) is shown in Figs. 1 and 2. Figures 1 and 2 show that significant amounts of hydrocarbons and hydrogen were formed during the processes. Propane ( $\text{C}_3\text{H}_8$ ) was decomposed to form various hydrocarbons with the relationship (ethylene > hydrogen > ethane > methane > propylene, where this relationship may change depending on operating conditions for different plasma reactors). All hydrocarbons and hydrogen increase with increasing applied power. Up to 6 to 7% of hydrocarbons and 3 to 4% of hydrogen were produced from the 10% propane mixed in nitrogen. The same figure shows that we obtain nearly 50% hydrogen selectivity; when defining hydrogen production selectivity from total hydrocarbon generated as:

$$\left[ \begin{array}{c} \text{Hydrogen Production} \\ \text{Selectivity} \end{array} \right] = \frac{[\text{Total Concentration Produced}]}{[\text{Total Hydrocarbon } C_x\text{H}_y \text{ by-product concentration } (x = 0 \text{ to } 3)]}$$

Nitrogen is used as a less reacting balance gas, where the optimum concentration of  $\text{O}_2$  with  $\text{N}_2$  balance can be obtained from air by pressure swing absorption (PSA). Argon or Helium can also be used. The advantage of using these balance gases is that they do not easily produce undesired compounds.

Energy efficiency of hydrogen production is shown in Fig. 3 as a function of specific energy density (SED: electric power input to reactor/gas flow rate), where the electrical power input from the secondary side pulse (0.1 to 0.3 W) was used in the present calculation. Energy efficiency increases with increasing

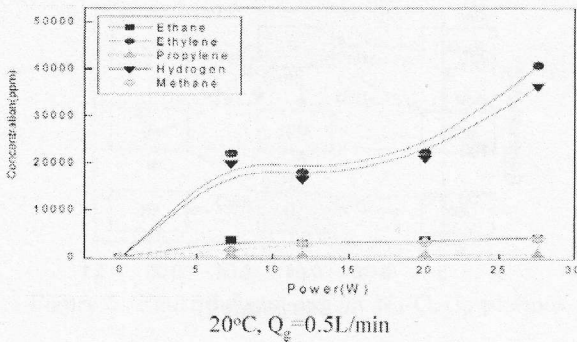


Figure 1. Hydrogen and hydrocarbon by-products generated by plasma as a function of applied voltage in  $N_2$ -10% propane systems (at  $20^\circ C$ ,  $Q_g = 0.5$  L/min,  $f = 50$  Hz) [19].

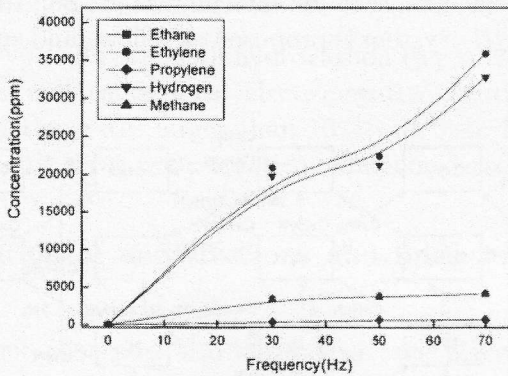


Figure 2. Hydrogen and hydrocarbon by-products generated by plasmas as a function of pulse frequency in  $N_2$ -10% propane systems (at  $20^\circ C$ ,  $Q_g = 0.5$  L/min,  $V_p = 10$  V) [19].

SED as expected. Approximately 219 g of hydrogen can be produced by 1 kWh of electricity, where this value is of an order of magnitude higher than the other non-thermal and thermal plasma methods (0.1 to 10 g  $[H_2]$ /kWh) [1-30].

### 2.3 $N_2-C_xH_y$ plasma chemistry

The mechanism of  $H_2$  formation is not well known. For the natural gas reforming processes, the thermal dissociation process is expected to occur at gas temperature ranges from 800 to  $1800^\circ C$ . Chang et al [32] proposed to replace thermal dissociation reactions of methane by electron impact initiated plasma reactions to produce this process under low gas temperature conditions ( $T_g < 100^\circ C$ ) and  $H_2$  formation was confirmed.



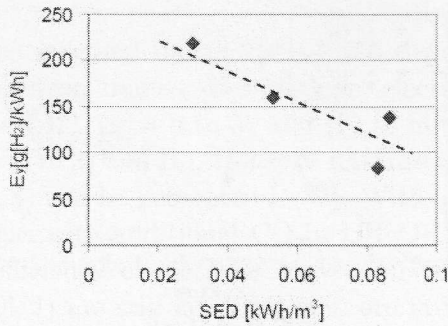


Figure 3. Energy efficiency of hydrogen production as a function of specific energy density (SED) (at 20°C, Q<sub>g</sub> = 0.5 L/min, f = 50 Hz) [19].

Based on available chemical reaction information [33-36], the plasma chemical kinetic model for N<sub>2</sub>-C<sub>x</sub>H<sub>y</sub> system is proposed as follows and summarized in Figs. 4 and 5.

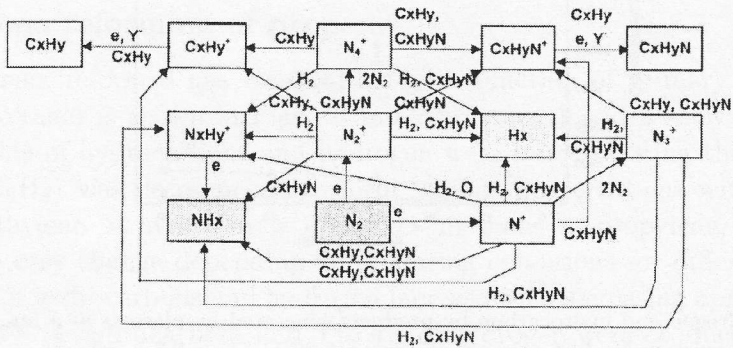
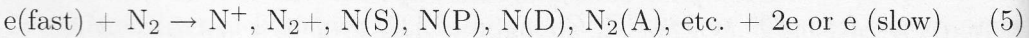


Figure 4. Ionic chemistry for N<sub>2</sub>-C<sub>x</sub>H<sub>y</sub> plasmas.

### Step I. Formation of active species

Since N<sub>2</sub> is the dominant composition (>90) in N<sub>2</sub>-C<sub>x</sub>H<sub>y</sub> systems, the plasma is generated only by electron impact processes with N<sub>2</sub> and form N<sup>+</sup>, N<sub>2</sub><sup>+</sup>, N<sup>\*</sup>, N<sub>2</sub><sup>\*</sup> species as follows:



where N<sup>+</sup> and N<sub>2</sub><sup>+</sup> also react with N<sub>2</sub> by three body reactions to form N<sub>3</sub><sup>+</sup> and N<sub>4</sub><sup>+</sup> in a high gas pressure plasma and excited or metastable N<sup>\*</sup> and N<sup>+</sup> can be also

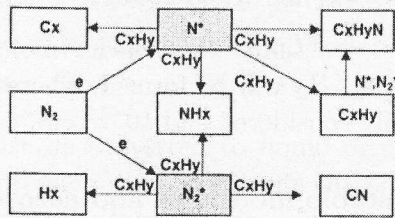


Figure 5. Neutral chemistry for  $N_2$ - $C_xH_y$  plasmas.

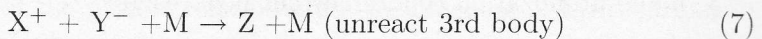
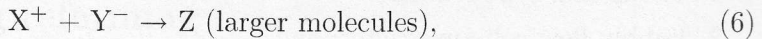
formed by a two-step electron ionization process.

## Step II. Formation of heavier positive ions and electronegative gases

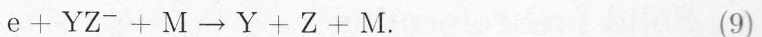
Electron and ions  $N_x^+$  will react with hydrocarbon to produce positive ions  $C_xH_y^+$ ,  $C_xH_yN_z^+$ ,  $NH_x^+$  as well as to form electronegative neutral species,  $H_x$ ,  $NH_x$ ,  $C_xH_yN_z$  where reaction rates larger than  $10^{-11}$   $cm^3/sec$  for two body reactions and  $10-30$   $cm^6/sec$  for three body reactions are considered in the present model.

## Step III. Formation of negative ions and initiation of recombination processes

Electron attachment processes also started to take place when electronegative gases were produced from step II and various negative ions will be formed from negative ion-molecule reactions.  $NH_x^-$ ,  $H_x^-$ ,  $C_xH_y^-$ ,  $C_yH_yN_z^-$  will be formed and these positive and negative charged species recombine to form larger molecules as follows:



where electron-ion volume recombinations normally form smaller molecules or atoms as follows:



All of these processes are summarized in Fig. 4 where the two and three body reaction rates larger than  $10^{-11}$   $cm^3/s$  and  $10^{-30}$   $cm^6/s$ , respectively, were only considered in this model.

### Step IV. Formation of heavier molecules

Based on nitrogen radical reactions, the heavier molecules can be generated. Again,  $\text{NH}_x$ ,  $\text{C}_x\text{H}_y$ ,  $\text{C}_x\text{H}_y\text{N}_z$ ,  $\text{H}_x$  will be formed, where the reaction rates larger than  $10^{-14}$   $\text{cm}^3/\text{s}$  were only considered and  $10^{-12}$   $\text{cm}^{-3}/\text{s}$  were marked in Fig. 5.

### Step V. Formation of stable molecules and aerosols

Atom-molecule and molecule-molecule reactions will take place and stable molecules will be generated. However, due to the ion-induced aerosol particles formations as well as molecule clustering reactions, some aerosol particles may be generated from the hydrocarbons. Based on these processes, it is clearly the plasma density that plays a major role in the generation hydrogen or formation of hydrocarbons.

## 3 Liquid fuel reforming

The objectives of the liquid fuel reforming are:

- cracking of liquid fuel to less viscous lower  $\text{C}_x\text{H}_y$  fuel, where the cracked liquid fuel is also expected to be a less polluting fuel, i.e. generating less soot formations [37-39];
- removal of pollutant elements from fuels such as S, N, Cl, etc.; and
- generation of gaseous fuels.

In order to maximize mass and heat transfers, the gas-liquid two-phase flow is used to maximize gas-liquid interfacial area and velocity as well as mixing. The most common types of reactors are [39]:

1. falling film type reactor;
2. bubbly flow column reactor; and
3. liquid atomization countercurrent flow reactor.

However, unlike ordinary gas-liquid flow, electrohydrodynamic phenomena will dominate flow on account of the existence of space charges and strong electric field [40, 41].

## 4 Solid fuel reforming

The objectives of solid fuel reforming are:

- conversion to high thermal value gaseous fuel normal from the mixture of solid fuel and steam;

- removal of metals and inorganics as fly and bottom ashes; and
- recycling of solid waste as gaseous or liquid fuels and construction materials – unreachable slugs.

Since solid phase must be converted to liquid or gas, only thermal plasma system can be used under reduction environments [40-42]. Typical thermal plasma pyrolysis products from waste automobile tires are shown in Tab. 1 [42]. Table 1 shows that a relatively high heating value fuel gas can be obtained from the thermal plasma pyrolysis of waste tires.

Table 1. Gas concentration and heating value from waste automobile tire [32]

Molecule	Concentration	Detection method
H <sub>2</sub>	5-20%	GC
CO	4-9%	GC, FTIR
C <sub>2</sub> H <sub>2</sub>	2-9%	FTIR
CH <sub>4</sub>	0.6-3%	GC, FTIR
C <sub>2</sub> H <sub>4</sub>	0.5-1%	FTIR
CO <sub>2</sub>	0.5-7%	FTIR
H <sub>2</sub> O		FTIR
N <sub>2</sub>	1-12%	GC
SO <sub>2</sub>	80-300 ppm	Detector
NO <sub>x</sub>	1 00-300 ppm	Detector
O <sub>2</sub>		GC
Ar		GC
Combustible gas	15-35%	
Combustion heat	4-7 MJ/m <sup>3</sup>	

## 5 Concluding reforming

The main objectives of the plasma reforming are the cracking of hydrocarbon to low C<sub>x</sub>H<sub>y</sub> or even to hydrogen level, and the thermal plasma technique demonstrated the feasibility of solid and liquid fuel reforming while the non-thermal plasma technique achieved energy efficient reforming of gaseous and liquid fuels. However, the energy efficiency of the plasma reforming depends significantly on the plasma system used. An optimization of the advance reactors and power supplies are required based on higher hydrogen yield or higher heating syngas yield with less toxic by-product formation.

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