On-Board Hydrogen Production System with Use of Plasma Discharges for Fuel Cell Transport:

The Effect of Discharge Pulse Width on Decomposition

プラズマを使用した車上における燃料電池用水素発生システム：放電パルス幅の効果

Y. Nishida*[1,2], H.C. Chaing[1] and T. C. Chen[1], T. Konishi[1,3] and C. Z. Cheng[1,2]
西田靖、江炫慶、陳資菁、小西匠、陳秋榮、

[1]Institute of Space and Plasma Sciences, National Cheng Kung University, Tainan 70101, Taiwan
[2]Research Center for Energy Technology and Strategy, National Cheng Kung University, Tainan 70101, Taiwan
[3] On leave from Electrical Engineering and Computer Science, Graduate School of Engineering, Iwate University, 4-3-5 Ueda, Morioka-city, Iwate 020-8551, Japan.

Abstract—Hydrogen production on vehicle without producing COx using plasma discharges working in high pressure condition over 1.5 atm. is under development. The original fuel for the system is methane (CH_4) or propane (C_3H_8). For those purposes mainly dielectric barrier discharge (DBD) non-thermal plasma system is employed with high voltage pulse of width 5-30 μsec and maximum amplitude 15 kV. Clear decompositions of hydrocarbons, ~70% of H_2 /CH_4 with input energy 110kJ/L (p=1.5 atm.) ~130 kJ/L (p=2.5 atm.) or 60% of H_2/C_3H_8 with 96 kJ/L at p=1.5 atm., have been observed. The decomposition rate depends on the input pulse width; each pulse is composed of multiples of fundamental 5 μsec pulse. The physical phenomenon of the pulse width dependence is shown to depend on the life time of plasma produced by each pulse.

Keywords—COx free hydrogen production; short pulse width dielectric barrier discharge; pulse width dependence

I. INTRODUCTION

Currently, clean energy source is one of the most important key issues, and the key technologies for clean and stable energy sources should be developed urgently. Most of the energy sources in the present world are fossil fuels and nuclear fission energy. However, consumption of the fossil fuels produces large amount of hazardous materials to the environment [1,2].

Hydrogen is expected to be one of the clean energy sources. The hydrogen burning has no major dirty output except just water vapor. Energy conversion efficiency to electricity with use of fuel cell can be expected to reach about ~ 60-80 %. This is one of the best clean energy sources with high conversion efficiency as an energy converter. The hydrogen feeding to the fuel cell, however, require a tremendously large amount of extra energy in addition to produce hydrogen by using popularly known methods, because most of the case hydrogen is fed to the consumption place by liquid or very high pressure state, i.e. reservation of hydrogen gas require a large amount of additional energy for making such state of hydrogen [3].

A new approach is to use plasma reforming. There are several techniques using plasma process such as arc plasma (ex. plasma torch) discharge, dielectric barrier discharge (DBD), corona discharge, catalyst pellets assisted packed-bed type discharge or microwave discharge system [1,4,5]. The original gas for decomposition is hydrocarbon.

In our decomposition system the following process is expected such as shown in Eq.(1) or (2);

$$CH_4 \rightarrow C + 2H_2 \tag{1}$$

$$C_3H_8 \rightarrow 3C + 4H_2 \tag{2}$$

The bond energies of hydrocarbons in each element are typically as follows [6],

$$C - H : 416\ \mathrm{kj/mol.} \approx 4.3\,\mathrm{eV} \tag{3}$$

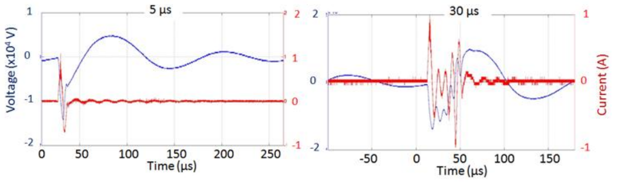
$$H - H : 436\ \mathrm{kj/mol.} \approx 4.5 \tag{4}$$

In the decomposition of hydrocarbon, the efficiency of decomposition may depend on the pulse width of applied high voltage pulse, so that in the present works, the effect of pulse width dependence is precisely investigated.

II. EXPERIMENTAL APPARATUS AND RESULTS

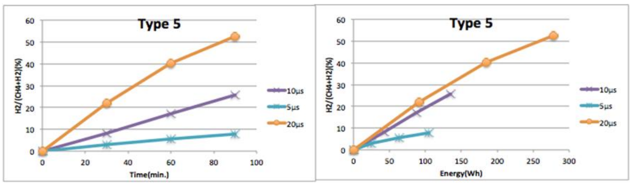
Experimental apparatuses including the system of discharge and detectors are shown in Ref.[1]. In the present works, the effects of pulse width on decomposition efficiency is performed precisely with high voltage pulse generator having 5 ~ 30 μs. Here, the pulse repetition rate of 4 kHz and the maximum amplitude of 13 kV are employed. All of the discharges were performed in high pressure conditions over 1.5 atmospheric pressures with use of pure hydrocarbon gases.

Examples of voltage (blue line) and current (red line) waveforms observed with change of pulse width 5 µs, and 30 µs are shown in Figs.1.(a) and (b), respectively. Here, the high voltage power source used in the experiments does not produce good waveform. If the pulse width is set-up more than 10 µs, the pulse is distorted by multiple oscillation. Typical example can be seen in Fig. 1(b), where a set-up voltage pulse width is square of 30 µs, which splits into about four of ~5µs oscillations and each short pulse makes discharges and thus for longer pulse width more plasma discharges take place to increase decomposition rate. Figure 2 shows the hydrogen production rate versus discharge time duration and input energy as a parameter of pulse width for 5 µs, 10 µs, and 20 µs. The results show that both hydrogen production rate and energy efficiency increase with the pulse width. To obtain 1 mole of Hydrogen, the production rate from methane requires energy approximately 0.33 kWh and the hydrogen production rate is (H_2/(CH_4+H_2))≃63% in 90 minutes.



(a) (b)

Fig. 1. The voltage (blue line) and current (red line) wave forms measured at the electrode in one cycle. Applied pulse width is (a) 5 µs and (b) 30 µs.



(a) (b)

Fig. 2. The hydrogen production rate v.s. (a) discharge duration and (b) input energy as a parameter of pulse width. Gas is methane.

Figure 3 shows an example of hydrogen production rate from methane versus the voltage pulse width. We can see that the 20 µs pulse width case gives the best hydrogen production.

To understand the results shown in Fig. 3, we consider the physical mechanism. Once the discharge occurs, free electrons are accelerated by the applied high electric field to collide with the methane molecules, resulting production of hydrogen molecules. Now, both methane and decomposed hydrogen molecules exist in the experimental chamber. Two types of reaction occur at the same time: decomposition and producing plasma state. The accelerated electrons get enough energy of about 1-20 eV to decompose methane gas molecules and making many hydrogen molecules.

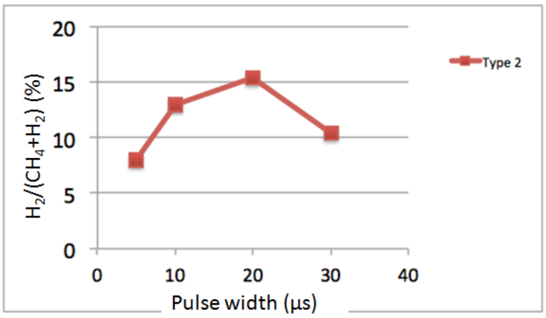


Fig. 3. Hydrogen production rate as a function of high voltage pulse width.

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