

Improvement of Plasma Reactor Performance for Hydrogen Generation

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Abstract : Research was performed to increase the efficiency of a plasma reactor for H₂ yield. In the preceding studies H₂ was increased by adding Ni as a transitional metal catalyst and TiO₂ as a photocatalyst. In these experiments, it was found that distilled water, discharge frequency, and electrode configuration had a significant impact on H₂ generation. A substantial amount of hydrogen yield was observed at 2 kHz of discharge frequency and 12 kV of applied voltage. Within this favorable discharge conditions, the weight rate of TiO₂ and Ni powders was investigated. Plasma phenomenon was measured by electrical, optical and acoustical devices. It was found that emitted light, electric current and acoustical signals acquired from the discharge demonstrated systematical correlation. Changing the electrode's configuration allowed discharge distribution along the perimeter of the electrode's tip, which increased the density of streamers and plasma energy loadings, as the value of inception voltage for the discharge propagation decreased.

Key Words : plasma reactor, photocatalyst, hydrogen production

1. Introduction

Direct water decomposition for hydrogen evolution is the process that requires energy. In this research study, pulsed power source was chosen as an energy supply for water splitting. This study is within the framework of hybrid pulse discharges [1]-[3].

2. Experimental setup

The schematic of the plasma system for hydrogen production is shown in (Fig. 1). The gap between the tip of the HV electrode and water surface was fixed at the previous optimal condition, 5 mm. The volume of the plasma reactor chamber was 120 cm³. N₂ was chosen as a carrier gas for plasma formation.

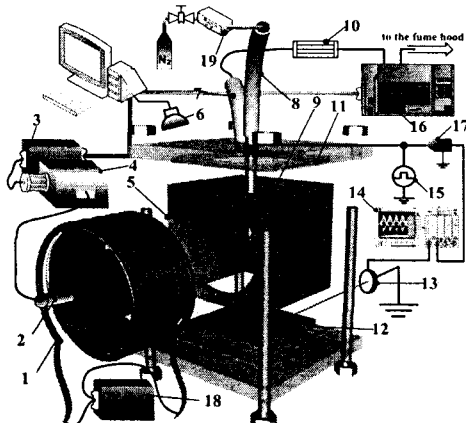


Figure 1. Schematic diagram of experimental set up

Needle type HV electrode used in our previous study [4] was replaced with the hollow type. Comparative characteristics in

discharge phenomenon using the two electrode types is demonstrated in Fig. 2. Ni and TiO₂, as water additives, were chosen to promote high rate of H₂ yield, in conjunction with recent research conducted by Sakae Takenaka et al. [5]. During the plasma reactor operation appeared acoustical signals. A double-channel FFT-based spectrum analyzer was used to measure acoustic responses.

3. Experimental results and discussion

Streamers were initiated by administering a wide range of applied voltage 7~12 kV according to the aqueous solution's composition and pulse frequency. It was identified that increasing frequency for distilled water, the value of the inception voltage decreased.

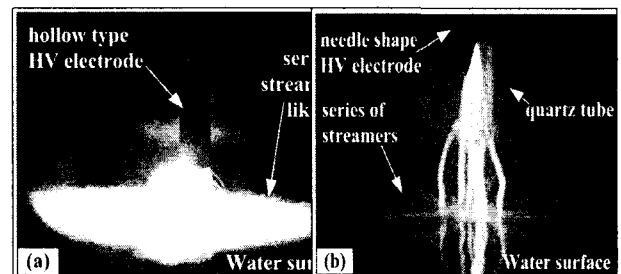


Figure 2. Difference in discharge phenomenon between hollow type (a) and needle type (b) HV electrode for 12 kV threshold voltage with 2 kHz discharge frequency, N₂ carrier gas with 0.2 L/min flow rate.

Threshold voltage (12~13 kV) was gained corresponding to the radical change in light emission from plasma and the discharge itself, see Fig. 2 (a). Fig. 3 plots typical waveforms of the pulse voltage and current in respect with Fig. 2 (a). In this experimental

study, strong compressional vibratory waves were observed within the audible range of sound (see Fig. 4). Signal peaks represent harmonics of the carrier wave, which is in part equal to the discharge frequency. It is shown that the highest strength (85 dB) belongs to the fundamental carrier wave, though in this case H_2 yield is at a low level. Acoustic spectrum for a high rate of H_2 production (Ni catalyst, 0.2 g) includes a dominant resonant acoustic wave of 8000 Hz frequency, corresponding to the 4th harmonic, with intensity nearing 83 dB. By varying discharge frequency (0.3, 1, 1.5 and 2 kHz), the resonant acoustic wave was also found to be within the 7000–8000 Hz range.

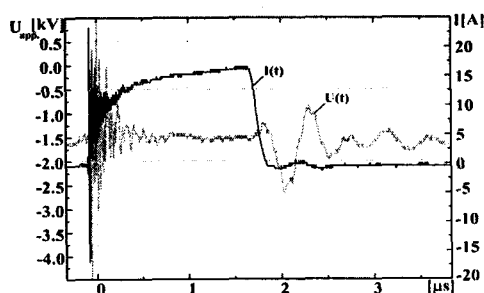


Figure 3. Typical waveform of pulse voltage and discharge current for pulsed discharge in N_2 environment; $U_{\text{appl}}=12$ kV, $f=2$ kHz.

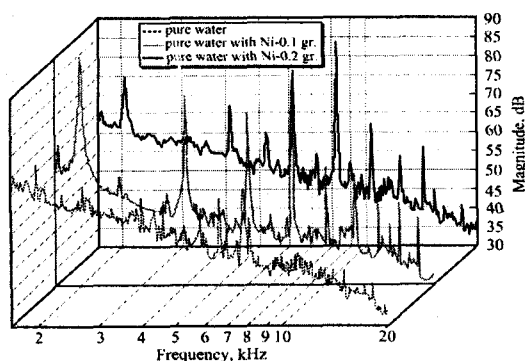


Figure 4. FFT acoustic spectrum at the 12 kV of applied voltage; 2 kHz discharge frequency; N_2 as a carrier gas; 5 mm gap distance; 1.5 mm diameter of the hollow type HV electrode.

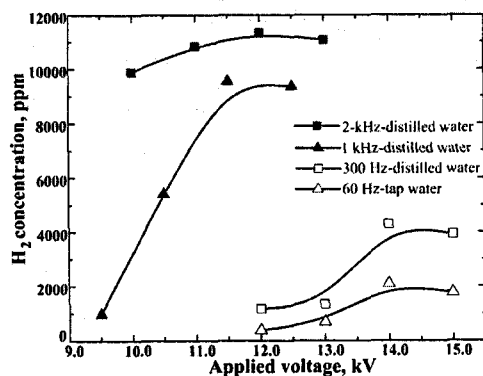


Figure 5. Effect of frequency on H_2 yield.

A steep increase of H_2 generation was observed in accordance with rising discharge frequency. Frequency effect along with a decrease in plasma loading, due to a fall in the optimal applied voltage (14–15 kV for low frequency and 12–13 kV for high frequency discharges) is demonstrated in Fig. 5.

4. Conclusions

The effects of discharge frequency, applied voltage and water additives on the production of H_2 in a hybrid plasma reactor were investigated. It was shown that H_2 production increases with the growth of applied voltage and discharge frequency. The highest H_2 concentration (12144 ppm) was obtained at 12 kV applied voltage and 2 kHz pulse repetition frequency. Streamer-like-arc discharges at the optimal condition of plasma reactor operation produced intense UV emission, which enhanced water decomposition by catalyst assistance. Substantial augment in H_2 evolution was achieved for Ni and TiO_2 additives with 0.2 and 0.3 grams respectively. Discharge itself emanated strong acoustic waves. Resonant acoustic frequency (8 kHz) was found to be in good agreement with the high rate of H_2 yield.

Acknowledgements

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References

- [1] P. Lukes, M. Clupek, V. Babicky, V. Janda and P. Sunka, Generation of ozone by pulsed corona discharge over water surface in hybrid gas liquid electrical discharge reactor, *J. Phys. D: Appl. Phys.* 38 (2005) 409416, 2005.
- [2] W. F. L. M. Hoeben, E. M. van Veldhuizen, W. R. Rutgers, G. M. W. Kroesen, Gas phase corona discharges for oxidation of phenol in aqueous solution, *J. Phys. D: Appl. Phys.*, 32 L133, 1999.
- [3] P. Lukes, A. T. Appleton and B. R. Locke, Hydrogen peroxide and ozone formation in hybrid gas-liquid electrical discharge reactors, *IEEE Trans. Ind. Appl.* 40 1 60-67, 2004.
- [4] J. Y. Park, J. S. Kim, N. V. Cong, S. B. Han, P. Kostyuk, S. H. Park and H. W. Lee, The co-effect of TiO_2 , Cu and Ni powders for enhancing the hydrogen generation efficiency using plasma method", *Proceedings of the 4th Asia-Pacific International Symposium on the Basics and Applications of Plasma Science and Technology* 157-163, 2005.
- [5] S. Takenaka, Y. Kobayashi and K. Otsuka, Formation of hydrogen through the decomposition of kerosene over nickel-based catalysts, *Energy & Fuels*, 18, 1775-1783, 2004.
- [6] M. Yousfi and M. D. Benabdessadok, Boltzman equation analysis of electron-molecule collision cross sections in water vapor ammonia, *J. Appl. Phys.* 80 (12) 6619-6630, 1996.