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Study on the Effects of Ultrasonic Wave for the Effective Hydrogen Generation by Electrical Discharge Plasma Process

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Abstract

The research was tried to investigate the hydrogen generation from water by the pulsed power plasma process. Hydrogen was generated by way of the electrical pulse power discharge process with the ultrasonic wave. The yield on the hydrogen generation was also studied with and without operating the ultrasonic generator, in which the applied high voltage was varied from 10 kV to 15 kV. Nitrogen and argon gases were used as working gases. As the results, the generation yield using the pure nitrogen gas is better than argon and mixed gases such as argon and nitrogen. Hydrogen concentration are significantly increased when the ultrasonic generator was operated with the electrical discharge simultaneously. It is increased with increasing the applied ultrasonic level as well.

Key Words : Plasma process, Hydrogen generation, Ultrasonic wave, Pulse power

1. INTRODUCTION

Researches on hydrogen production over several decades have performed for the efficient and economic generation[1-3] Especially, hydrogen becomes important energy source to our life due to its noble characteristics such as clean, safety, renewable, and abundant element on the earth. In addition, hydrogen is believed to be an alternative means for the near future energy source. Several generation methods for hydrogen have been developed. In particular, as one of those methods, the approach using electrical discharge playes an important role in semiconductor processes, the high efficiency lighting development, arc welding machines, plasma enhanced chemical processing, and toxic substances treatment as well as the hydrogen generation[9-12]. However, the direct decomposition of water molecules for the hydrogen generation is not easy. Usually, hydrogen can be obtained by the electrolysis and the pyrolysis processes[3-5]. Nowadays, many researchers have attempted to apply plasma technology for the efficient hydrogen production in water. It is said that ultrasonic wave has a various number of applications, especially on the field of environment such as surface cleaning, organic contaminants removal etc.

However, there are not so much related works dealing with the ultrasonic effects on the field of the hydrogen generation using the plasma technology. In this paper, it was investigated to enhance the efficiency on the hydrogen generation using a ultrasonic wave generator.

The non-thermal plasma reactor with a single needle is designed and operated with the pulsed high voltage power of 60 Hz frequency. Besides, the effect of ultrasonic generator, the discharge power, and the applied voltage on the hydrogen generation efficiency are investigated in details.

The discharge power was calculated by multiplying the input applied voltages with the

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corresponding discharge current. And the mechanism on the hydrogen generation using this reactor is also discussed.

2. EXPERIMENTS

2.1 The plasma reactor

Figure 1 shows schematic of the plasma reactor used. This discharge system consists of the reactor chamber made of acrylic tube with the dimension of 30 mm in height and 60 mm in inner diameter, and quartz tube for inserting the inner high voltage needle electrode, which is made of stainless steel with the diameter of 2 mm.

In addition, the ground electrode is made of copper film and it is dipped into water to become grounding electrode. The gap between the tip of the inner high voltage electrode and the lowest point of quartz tube was fixed at 5 mm. The gap between the lowest point of quartz tube and the water surface is also constant at 5 mm.

Figure 2 shows schematic diagram of the experimental setup. The experimental setup consists of the high voltage pulsed generator, MFC (mass flow controller) that can control the amount of carrier gas, watt meter to estimate the consumption power when discharge occurred, oscilloscope to record voltage waveform through a voltage divider and discharge current, and GC system (Gas Chromatography - Younglin Instru. Co., Model 600D) for estimating the hydrogen concentration and analyzing by-products. The detector type of this GC system is PDD (pulse discharge detector) which has the sample loop of 2mL with the diameter of 1.6 mm.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1 Discharge characteristics

In this research, we have performed two types of experiments. The first type is about the effect of the electrical discharge on the hydrogen generation efficiency by the only plasma reactor without using ultrasonic wave generator. The second type is to investigate co-effects of the electrical discharge and the ultrasonic generator on the hydrogen efficiency under the same conditions of other parameters.

In this work, the ultrasonic wave is considered as a promoter to increase the efficiency of the hydrogen generation of the plasma reactor.

For both of these above experiments, the same conditions that support the discharge phenomena are setup and maintained stable. Nitrogen and argon are used as the carrier gases and their flow rates are constant at 200 mL/min in the case of only N_2 or, Agon, and 100 mL/min for each gases in the case of using mixed gases (volume ratio = Ar : 50 %, N_2 : 50 %). And the discharge time is 10 minutes for every applied voltage levels because the saturation of hydrogen in the flow line takes 10 minutes. The pulsed power is used for generating the strong electrical discharge.

In the case of without using ultrasonic generator with N_2 carrier gas, the discharge voltage is changed from 12 kV to 15 kV. When the ultrasonic generator is used, the discharge started from 11 kV. Which is lower than the former cases. The reason is that the ultrasonic wave effects can enhance the initiation of electrical discharge. This effects makes the stronger electrical discharge at the same applied voltage as well as increase the hydrogen efficiency.

However, when the applied voltage and the applied ultrasonic wave levels are increased at the same time, the stable discharge are limited to 15 kV. At this point, discharge phenomena are suddenly decreased and changed to very unstable above that applied voltage, which occurred in any cases without or with using ultrasonic generator. Under this change of discharge condition, the hydrogen concentration decreased as low as 552 ppm and 129 ppm. It corresponds to the applied ultrasonic wave level 2 and 3, respectively. The discharge power on the applied voltage is showed in the following Fig. 3.

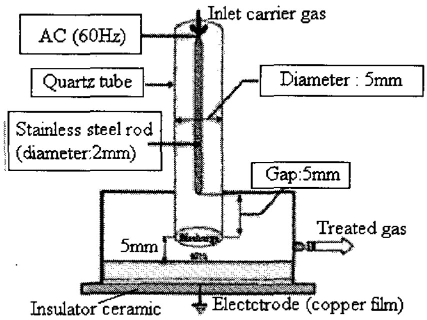

Fig. 1. Schematic diagram of the plasma reactor.

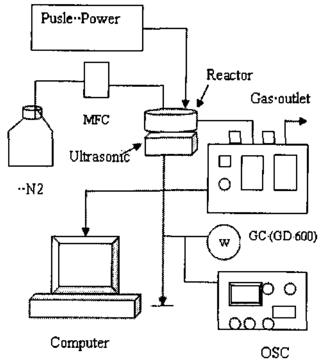

Fig. 2. Schematic diagram of the experimental setup.

3.2 Experimental setup

In this Fig. 3, the discharge power in all of cases are linearly increased with increasing the applied voltage. However, the discharge power with using the ultrasonic wave generator is a little bit high compared to without using the ultrasonic wave generator. From this results, it is assumed that the ultrasonic wave generator is affected to the discharge.

In the case of the mixed gases (N_2:50 % and Ar: 50 %) as carrier gases, the discharge phenomena are early taken place from about 10 kV. Therefore, the applied voltage is varied from 10 kV to 13 kV. The discharge phenomena are stable but not strong in this gas composition. It is also limited to 13 kV due to changing the discharge state into unstable.

When the ultrasonic wave generator was applied, the discharge phenomena are more strong and stable until they reach to 13 kV. It results from the higher discharge power than the case of without using ultrasonic wave generator as indicated in Fig. 3.

To enhance the hydrogen generation efficiency, the effects on the change of the water quantity inside the reactor are investigated with using the mixed carrier gases. When the water volume inside the reactor is 15 mL, it is about 5 mL high compared to the previous one. The stable discharge is obtained in the applied voltages from 10 kV to 13 kV. Especially when compared with the case of 10 mL, the discharge power is much lower. The discharge power is increased

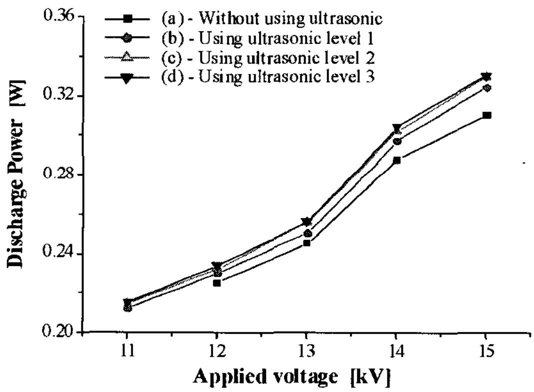

Fig. 3. The relationship between the power consumption and the applied voltage.

with increasing the applied ultrasonic wave levels at the same applied voltage. Nevertheless, this setup do not enhance the efficiency of the hydrogen generation as shown in the following Fig. 4.

3.3 Effects of the applied voltage and carrier gases

When an sufficient electrical field (DC, AC or pulse) for discharge generation is applied to gases, the energetic electrons generated by electrical discharge transfer their energy to the gas molecules by collision, which leads to the excitation, attachment, dissociation or ionization

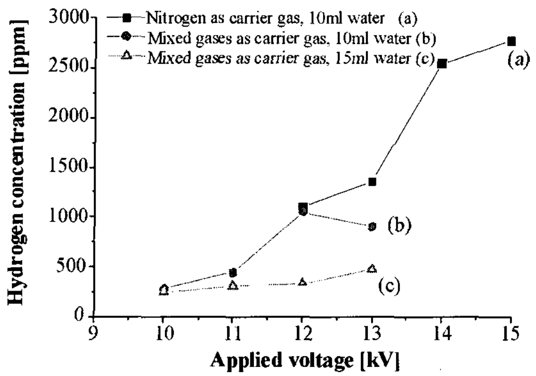

Fig. 4. The hydrogen concentration in the cases of without using ultrasonic wave generator.

of the molecules[4]. Therefore, the chemical reactions following by electrical discharge would be taken place inside the discharge reactor as discussed in [4-8].

The hydrogen generation efficiency are examined under the different carrier gas types such as nitrogen gas, the mixed gases of nitrogen and argon. In addition, the investigation about the effects of the water quantity is carried out. In the above Fig. 4, the effects of the applied voltage on the generated hydrogen concentration was showed.

The first type of experiment is done with the input applied voltage varying from 12 kV to 15 kV without using the ultrasonic generator. Nitrogen is used as the carrier gas and the water volume inside the reactor is 10 mL. The concentrations of the hydrogen are observed as varying from 1105 ppm to 2764 ppm, respectively.

The second type of experiment is done with the input applied voltage varying from 10 kV to 13 kV without using the ultrasonic wave generator and the mixed gases is used as the carrier gas, and the water volume inside the reactor is 10 mL. As showed in Fig. 4, the results show that the hydrogen concentration is increased with increasing applied voltage. It are varied from 276 ppm to 1105 ppm from 10 kV to 12 kV.

The discharge is limited as the applied voltage is increased up to 13 kV with the hydrogen concentration decreased about 525 ppm due to the unstable in the discharge conditions.

In the third cases, the water inside the reactor is increased to 15 mL. Under the effect of the pulsed power discharge in the range of the applied voltage from 10 kV to 13 kV, the hydrogen gas is very low. The hydrogen concentration is varied from 248 ppm to 470 ppm with the applied voltage from 10 kV and 13 kV, respectively.

Comparing the above three cases, the discharge power and the hydrogen generation efficiency are strongly depended on the applied voltage and carrier gas. We obtain the best results in the case of using nitrogen carrier gas, while the efficiencies in other cases are rather low. Even when water quantity increases from 10 mL to 15 mL in mixed gases, the hydrogen concentration does not increase.

The best hydrogen concentration is created in the case of using nitrogen carrier gas and water of 10 mm. This result seems to be related with the characteristic of carrier. While in the case of using the mixed gases, the efficiency is very low even at the same water quantities of 10 mL. The discharge also limited up to 13 kV. The electrical discharge is rather weak, especially with the volume of the water of 15 mL.

Plasma energy is likely that electrons accelerated by electronic field are consumed to excite argon gas not to dissociate water molecules as the case of nitrogen gas. Argon gas has its own characteristic of an inert gas. Therefore, it can not react with the oxygen molecules as well as others oxygen radical generated by plasma as well as nitrogen. The results of this process is that these kinds of excited species such as oxygen and oxygen radical will recombine with the hydrogen radical (H.), which is the main source to form the hydrogen gas. While nitrogen gas has this ability which it can react with oxygen into NO_2 or NO. It is contributed to the process for preventing the recombination among hydrogen atom and oxygen molecules and radicals.

This is the reason of this difference among these types and the chemical reactions under the different nitrogen gas and argon. It might cause the difference of the hydrogen generation. The discharge power and the hydrogen generation efficiency in argon carrier gas is not high.

3.4 The effects of the ultrasonic wave

3.4.1 Carrier gas effects on nitrogen gas

To investigate the effects of ultrasonic wave generator. The investigation about the hydrogen generation efficiency related to the three carrier gases are conducted. For the first type, other conditions are maintained as the same as the previous conditions except the present of ultrasonic wave. The correlation between the increase of the hydrogen generation efficiency and the increase of the ultrasonic wave is shown in the following Fig. 5.

The ultrasonic wave enhancement is controlled by adjusting the applied level from low level to higher level. The main difference of those level are electrical current supplied to ultrasonic wave generator according to each cases.

Within the applied voltage varying from 11 kV to 15 kV, the discharge phenomena are changed very clearly as the applied ultrasonic wave level varied from level 1 to level 3.

Under the effects of the ultrasonic wave, the consumption power are slightly increased. The ultrasonic wave shakes the reactor and creates the waves on the surface of the water inside the reactor. The higher of the applied ultrasonic wave level is sullied, the stronger vibrated water surface is created. It is also creating the turbulent of the water movement inside the reactor.

Thus, at all of the applied voltage, the discharge is much stronger with increasing the applied ultrasonic wave level. The maximum hydrogen generation is obtained as almost at 14 kV in cases of level 2 and 3. It show about 3589 ppm and 3565 ppm, respectively. In the case of the level 1, the hydrogen concentration is about 3510 ppm at 15 kV. However, the discharge at the two higher applied voltage in cases of level

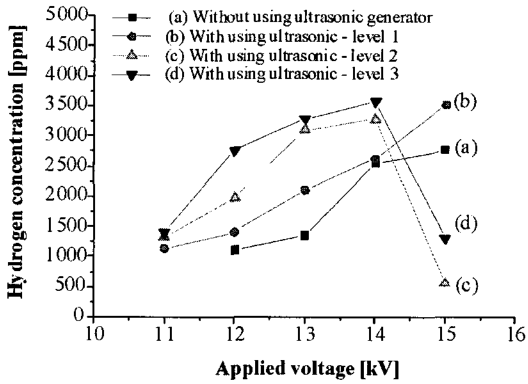

Fig. 5. The comparison of the hydrogen concentration in cases of using ultrasonic wave generator under N_2 and 10 mL water quantities.

2 and level 3, the hydrogen concentration is decreased into 552 ppm and 1299 ppm respectively.

The main reason leading to this fact is that the electrical discharge is unstable and significantly weak largely due to the strong effect of the ultrasonic vibration in the high applied voltage. From Fig. 5, the range of the applied voltage in case of with operating ultrasonic wave generator is widened to the lower level. It means that the electrical discharge is generated easily in comparison with the case without using that. Besides that, it is very clear that the hydrogen concentration at about 14 kV is much higher with the help of the ultrasonic wave enhancement.

3.4.2 Carrier gas effects on the mixed gas

The correlation between the effects of the ultrasonic wave enhancement and the hydrogen concentration in the case of using mixed gases are shown in the Fig. 6.

In this experimental conditions as previous experiments, the carrier gas is changed into the mixed gases of the nitrogen and argon gases. The water volume inside the reactor is the same as the previous one of 10 mL. The range of the applied voltage is changed from 10 kV to 13 kV.

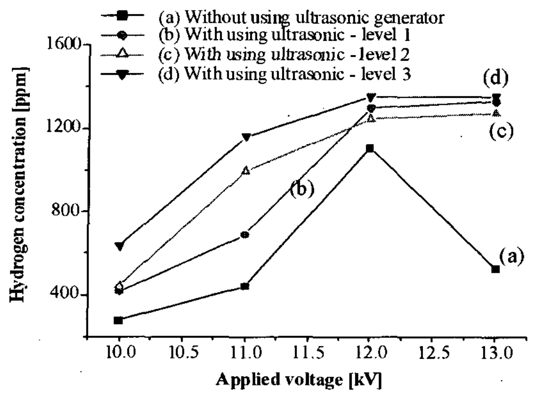

Fig. 6. The comparison of the hydrogen concentration in cases of without and with using ultrasonic wave generator under the mixed gas and 10 mL water quantities.

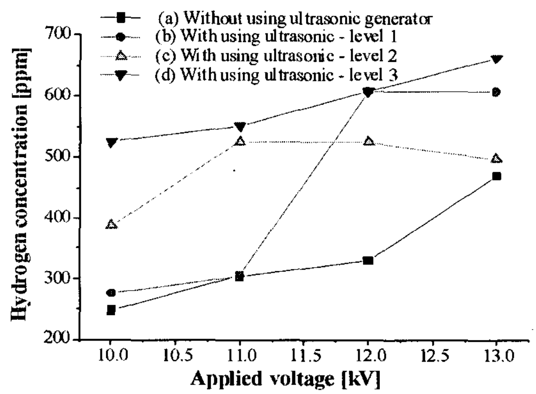

Fig. 7. The comparison of the hydrogen concentration in cases of without and with using ultrasonic wave generator under the mixed gas and 15 mL water quantities.

The applied ultrasonic level is the same as the above type varying from level 1 to level 3.

Under these conditions, the discharge power is rather weak. The stable discharge is limited to 13 kV. Without using ultrasonic wave generator, the hydrogen concentration is increased to 1000 ppm at 12 kV, but it is decreased to 52 5ppm with increasing the applied voltage of 13 kV.

In other cases, when using the ultrasonic wave generator, the discharge phenomena are much stronger, it results in the increase of the hydrogen concentration.

In Fig. 6, in all cases of this type, the hydrogen concentrations are not so much high in comparison with only nitrogen carrier gas showed in Fig. 5.

The peak point which is observed at 12 kV and 13 kV with ultrasonic wave level 3 is about 1354 ppm. The same situation compared to nitrogen carrier gas is occurred. The electrical discharge is limited to 13 kV in the high level 3 of ultrasonic wave generator.

3.4.3 The mixed carrier gas effects with changing water quantities

The correlation between the hydrogen concentration and the ultrasonic wave effects in the case of using mixed carrier gas are shown in the Fig. 7.

In this case, all of the experimental setup conditions are the same as the previous experiment, except for increasing water quantities into 5 mL. The electrical discharge is not changed largely but the discharge power is lower and the hydrogen concentration is also very low.

From Fig. 7, the hydrogen concentration in the case of without using ultrasonic wave is very low. The peak concentration is detected at the applied voltage of 13 kV and the applied ultrasonic wave level 3 and the hydrogen concentration is 663 ppm. This level of hydrogen concentration is considered as a very low. Nevertheless, the effects of the ultrasonic wave on the electrical discharge and the hydrogen generation efficiencies are clearly acknowledged.

4. CALCULATION OF ENERGY YIELD

From the whole of these works, it could be said that the role of the nitrogen carrier gas and the ultrasonic wave for enhancing the hydrogen concentrations are more effective than other cases. To understand more details about these, the energy yields of hydrogen generation are calculated as showed in the following Fig. 8.

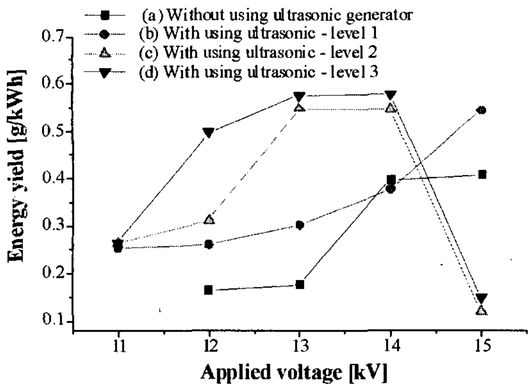

Fig. 8. Energy yield in cases of using N_2 carrier gas with ultrasonic wave generator under 10 mL water quantities.

In the above Fig. 8, we can see that, the energy yield is increased with applying the ultrasonic wave generator. The higher of the applied ultrasonic wave level is used, the better energy yield is obtained.

These phenomenon are limited when the applied voltage goes up to 15 kV at level 2 and level 3 of the ultrasonic wave generator. The reason is that the discharge is limited to that applied voltage and it is unstable over that applied voltage. However, the best energy yield is obtained at the applied voltage of 13 kV and 14 kV and with ultrasonic wave level 3 and it is about 0.5758 [g.kWh], which corresponds to the applied voltage of 14 kV.

5. CONCLUSION

From the these results obtained in the above experiment, it could be concluded as follows.

(1) The optimum conditions in these researches for producing hydrogen are: using the nitrogen gas as carrier gas with the flow rate of 0.2 L/min and using the ultrasonic wave level 3 (3565 ppm) and the applied voltage as 14 kV.

(2) The ultrasonic wave has a strong effect for enhancing the discharge power and increasing the hydrogen generation efficiency. The suitable range of applied voltages could be shortened from 12 kV to 14 kV instead of from 11 kV to 15 kV when the ultrasonic wave generator is used.

(3) The applied voltage level is strongly affected to the hydrogen generation efficiency. The hydrogen concentration is increased when the applied voltage is increased. The discharge gap and water quantities play an important role to obtain the optimum state of the discharge phenomena and power as well.

(4) The nitrogen gas is more suitable carrier gas compared with argon and the mixed gases. And the best energy yield is obtained at the applied voltage of 13 kV and 14 kV and ultrasonic wave level 3 and it is about 0.5758 [g.kWh] at the applied voltage of 14 kV.

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