Structural Properties of Dielectric Barrier Reactor with Hole (DBH) for CF_4 Decomposition

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In this paper, the CF_4 decomposition efficiency is investigated for three simulated plasma reactors that are needle plate reactor, metal particle reactor, and dielectric barrier reactor with hole (DBH). The CF_4 decomposition efficiency by DBH is much better than that by needle plate reactor or metal particle reactor. When applied voltage is increased up to the critical voltage for spark formation in the all reactors, the CF_4 decomposition efficiency is increased. The CF_4 decomposition efficiency in needle plate reactor and metal particle reactor is about 12% and 22% respectively at applied voltage of 23 kV (consumption power: 110 W) and CF_4 concentration of 500 ppm, however, the CF_4 decomposition efficiency is more than 95% in case of DBH. DBH should be much better than two reactors investigated for CF_4 decomposition.

Keywords : Needle plate reactor(NPR), Metal particle reactor(MPR), Dielectric barrier reactor with hole (DBH), CF_4, Decomposition

1. INTRODUCTION

In the last twenty years, semiconductor industries have been getting bigger and very important technology in a recent industry field. CFC (Chlorofluorocarbon), which is used for etching process in the semiconductor manufacturing process, CVD (chemical vapor deposition), and dry cleaning, is realized as the one of materials to destroy an ozone layer, so that scientists have tried to decrease the use of CFC in a semiconductor industry or remove it by several technologies.

PFC (perfluorocarbon) emission to atmosphere makes the surface temperature of the earth increased because the lifetime of PFC is really long to compare with other gases. The regulation plan about using PFC, which will be started from 2010, has already established internationally. Therefore, it has to be studied to find the suitable treatment methods of PFC and to develop alternative materials [1-3]. At present, thermal plasma and non-thermal plasma is generally used for PFC treatment technologies [4]. The PFC treatment technology with thermal plasma needs high power, so that economical efficiency and practicality are very bad. On the other hand, PFC treatment technology with non-thermal plasma could be good economically. There are many kinds of non-thermal plasma technology; SPCP (surface discharge induced plasma chemical processing), PPCP (pulsed plasma chemical processing), Ferro electric packed bed corona, and RF (radio frequency) plasma [5-7].

In this paper, the decomposition efficiency of CF_4 and the discharge type are investigated for three non-thermal plasma reactors at different conditions. And effect of reactor structure and discharge type on CF_4 decomposition is analyzed

2. EXPERIMENT

There are three types of reactors that are used in this experiment. N_2 is used to dilute CF_4 of 50% balanced by N_2 and AC high voltage is supplied for discharging gases. The figure 1 shows schematic diagram of the experimental setup. As showed in this figure, the experimental setup compose of the equipment of high voltage supply which is made by Pulse Inc., MFC (mass flow controller) which is possible to control the flow of CF_4 and N_2, gas mixing chamber which mix the pure gases controlled by MFC, gas cell for measuring FTIR (fourier transform infrared spectroscopy), and a wattmeter. All of these are located in Fume Hood.

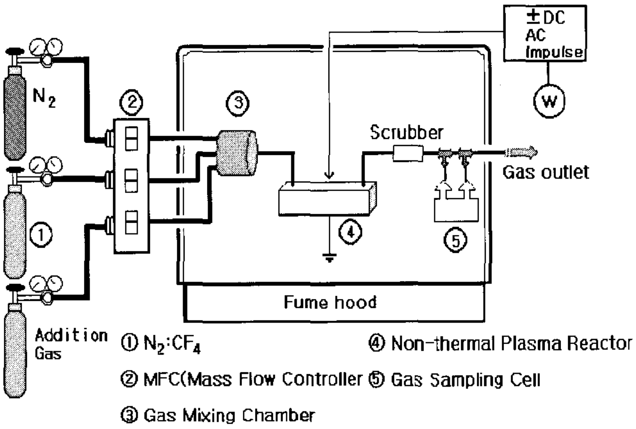

Fig. 1. Schematic diagram of the experimental setup.

The gas controlled by MFC is injected into non-thermal plasma reactor through the mixing chamber. The gas treated by non-thermal plasma is piled up into gas cell for FTIR measurement. We calculated decomposition rate of CF_4 with the intensity of FTIR spectra. When the flow rate of mixing gas (N_2:CF_4) is fixed to 2 ℓ /mm, removal characteristics of CF_4 is measured in case of the type of reactors, applied voltage, and gas concentration. In each time after experiment, N_2 gas is flowed for 5minutes to remove remained gases in plasma reactor and gas cell and new experiment is done

3. RESULTS AND DISCUSSION

3.1 Discharge characteristics

Three reactors are used in this experiment to investigate CF_4 decomposition as the structure of non-thermal plasma reactors, such as needle-plate reactor, metal particle reactor, and dielectric barrier with hole (DBH).

CF_4 (tetrafluoromethane) gas is belonging to PFCs that have very strong bonding energy comparatively. Therefore, glow or streamer discharges that have weak density are very difficult to decompose CF_4 gas. And CF_4 gases are adsorbed in solid surface because CF_4 gas has electron coherence [8], and then plasma to destroy this gas is to be very uniform and strong.

Figure 2 shows a form of needle-plate electrode type reactor. The reactor consists of a needle (length: 1.5 mm, ø: 1mm) and a copper plate (60× 70 mm). The gap between the needle electrode and the grounded plate electrode is 5 mm, the distance between the needles is 4 mm.

In needle-plate type reactor, uniform plasma is generated in whole reactor. Injected gas is passed through plasma field. Therefore, it is possible for all gas molecules to get plasma energy in this type reactor, but when gas flow increase, plasma is locally disappeared.

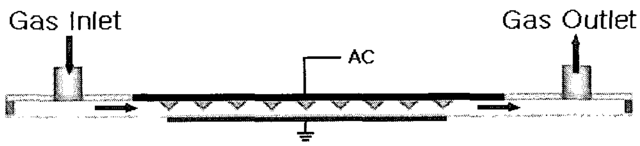

Fig. 2. Needle-plate electrode type reactor.

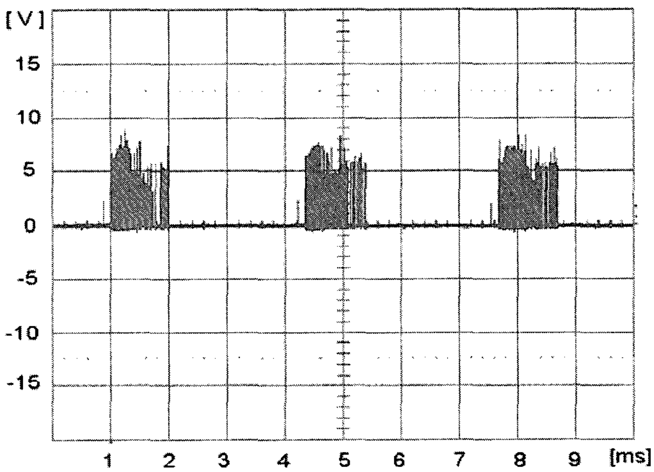

Fig. 3. Discharge oscillogram of needle-plate type reactor [15kV].

Figure 3 shows discharge oscillogram of needle-plate electrode type reactor. The typical streamer propagates from needle electrode to plate electrode with increasing voltage. Also, streamers only propagate to the positive.

Figures 4 shows metal particle reactor. Metal particle type reactor generates arc-like plasma compared with needle-plate electrode type reactor.

Arc discharge is very useful discharge type for decomposition processing of CF_4 with strong bonding energy because arc discharge has higher energy density than glow or streamer discharge.

Discharge of metal particle type reactor have characteristic such as thermal plasma in spite of non-thermal plasma. Arc discharge might effectively decompose CF_4 gas because arc discharge can transmit high-density energy to gas instantaneously compare with glow discharge or streamer discharge. Metal particle type reactor could have higher decomposition rate than that of needle-plate electrode type reactor.

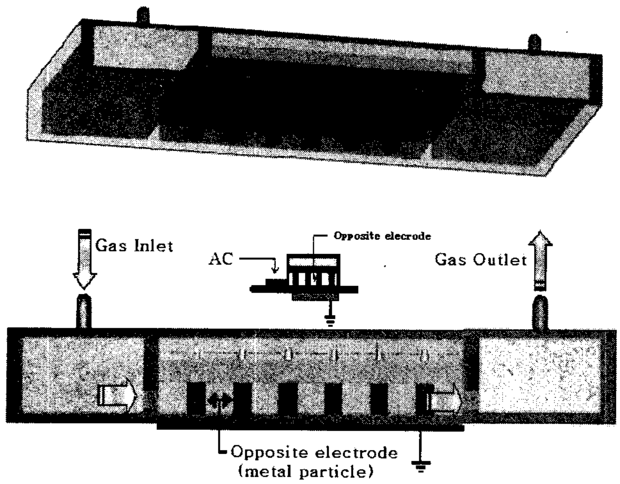

Fig. 4. Metal particle reactors.

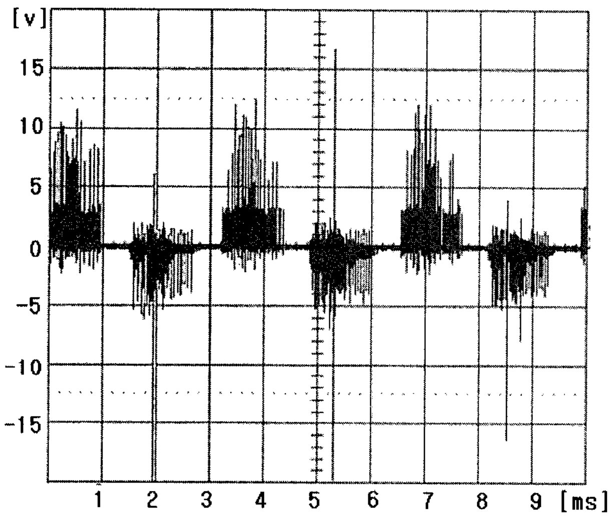

Fig. 5. Discharge oscillogram of metal particle reactor [15kV].

Figure 5 shows discharge oscillogram of metal particle type reactor. In more than 15 kV, it is appeared that the peak of pulse rises very irregularly. This peak is a high-density streamer that moves through the surface of reactor from one metal-particle to other metal-particle. At this time the gas decomposition rate is raised. Therefore the gas decomposition is greatly influenced by irregular pulse. In case of metal-particle reactor, the density of pulse is higher than that of hole type reactor, but this reactor has defect that the plasma density is lower than that of hole type reactor in the bulk area of the reactor and non-discharge area in the bulk increase because the pulse are moved through the surface of reactor from metal-particle to other metal-particle with only surface discharge type.

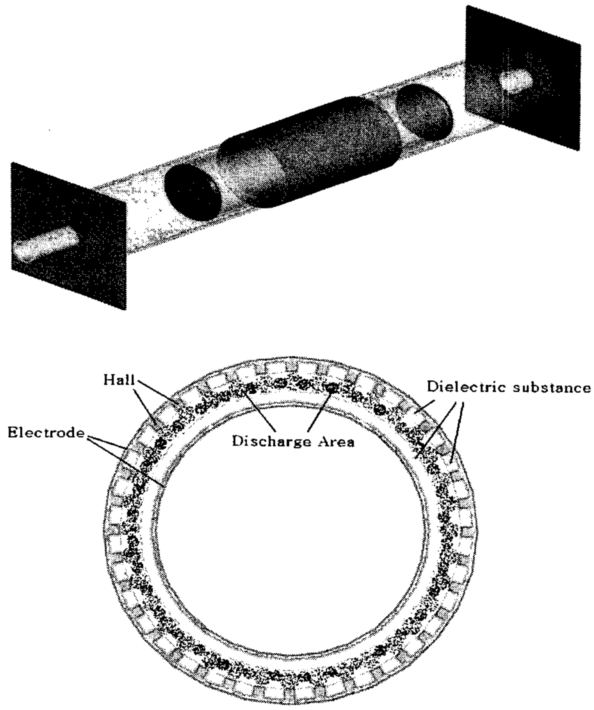

Fig. 6. The dielectric reactor with hole.

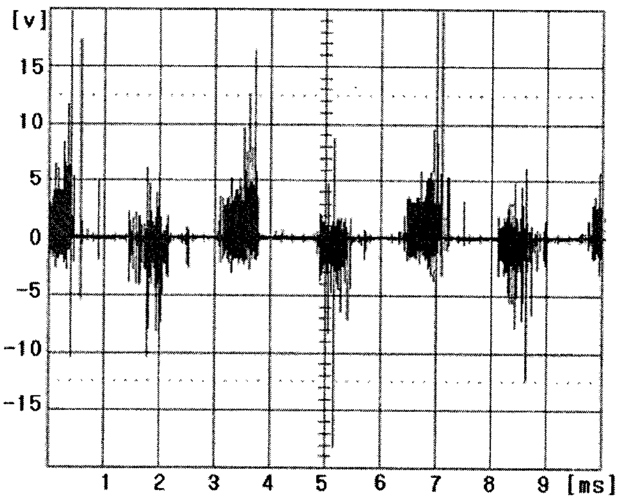

Fig. 7. Discharge oscillogram of dielectric reactor with hole [15kV].

The reactor such as figure 6 is made to improve decomposition efficiency by solving the problems of metal particle reactor.

In the case of DBH, the generated plasma is weaker than arc discharge, however stronger than glow discharge or streamer discharge. In other words, high-density streamer could be generated and we would call this discharge type arc-like discharge with high-density streamer. Therefore, it is thought that the reactor could reduce non-discharge area that is problem in metal particle reactor.

Arc-like streamer discharge is occurred at hole edge of dielectric barrier in DBH, which have many small hole in dielectric barrier that is put between electrodes. And plasma with strong streamer enough to destroy CF_4 can be seen in this reactor.

Figure 7 shows the oscillogram of discharge current in the DBH. In case of metal-particle reactor, the pulse is regularly generated to positive and negative direction. However in case of DBH, the similar pulse is generated to positive and negative direction. And high-density plasma is generated though the bulk area in reactor. Average current is smaller than that of the metal-particle reactor.

3.2 CF_4 Decomposition characteristic

The figure 8 shows decomposition rate of each reactors at 500 ppm and 2 ℓ /mm. The decomposition rate of metal particle reactor and needle-plate type reactor is at most below 20 % at 24 kV, but it of DBH is 75 % at applied voltage of 6 kV. When voltage is 23 kV, dielectric barrier reactor with hole have the decomposition rate of more than 95 %, in case of 100~400 ppm.

The decomposition rate of DBH as function of CF_4 concentration is showed in figure 9. CF_4 decomposition slightly increases as CF_4 initial concentration blow 100 W. But when consumption power is more than 100W the decomposition rate is rapidly increased. Plasma form below 100W has almost streamer form and high-density streamer and arc are generated at the same time over 100W.

According to consumption power, the decomposition rate doesn't monotonously increase. When arc area increase, high density streamer area decreases and non-discharge area is occurred, and then the decomposition rate decreases. Therefore the structure of reactor is very important and suitable plasma must be generated for good decomposition.

When arc is generated, electrical breakdown is necessarily occurred with increasing consumption power. In case of DBH pre-breakdown phenomenon is happened at 110 W. When gas concentration is 900 ppm, it is difficult to CF_4 gas decomposition by the high-density streamer and the decomposition rate is about 60 % at 10 W. But because some arc can improve the gas decomposition ability, decomposition rate increases to 70 % over 100 W. As voltage approaches to pre-breakdown voltage, the decomposition rate increases and it obtains to 85 % just before pre-breakdown. These results indicate that only the high-density streamer is difficult to remove high concentration gases. Arc-like plasma with high-density energy is very effective to decompose CF_4.

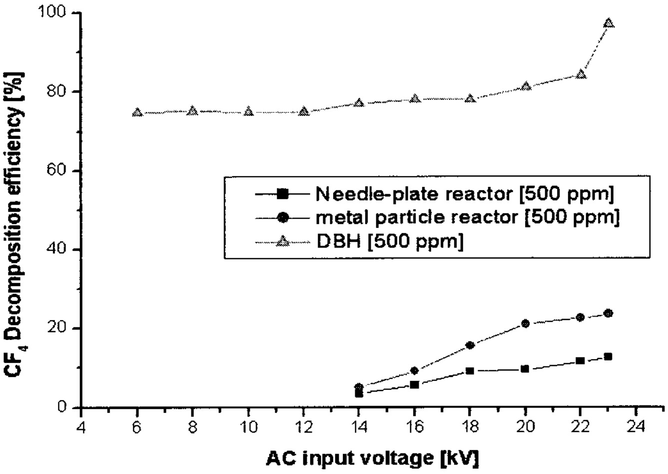

Fig. 8. Decomposition efficiency as function of CF_4 concentration in each a reactors [500 ppm].

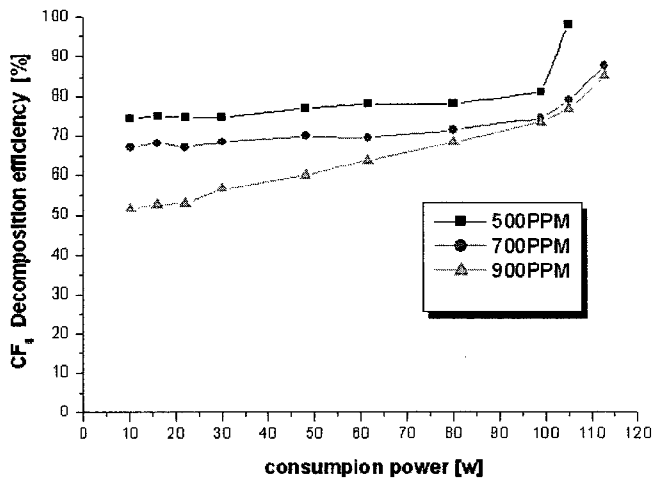

Fig. 9. The decomposition rate of CF4 gas by consumption power.

3.3 FTIR Spectra

The FTIR are shown in figure 10. As shown in these Figure, the peak intensity of the CF_4 decrease with increasing of applied voltage, however it of CO_2 increases.

Expected byproducts such as CO_2, COF_2 and CO are produced by the following chemical reactions between active radicals and ions [9-11] and the conversion process of CF_4 separated by the two steppes in figure 11 [12-15].

In the first step, CFx (CF_3, CF_2, CF) radical are created by effect of the electron collision. In the second step, CO_2 and COF_2 are created by reactions of O atom and CFx radical. However, trace byproducts predicted by the present model such as F_2, FOx, FO, and COF_2 is not observed, trace CO_2 is increased as a function of applied voltage.

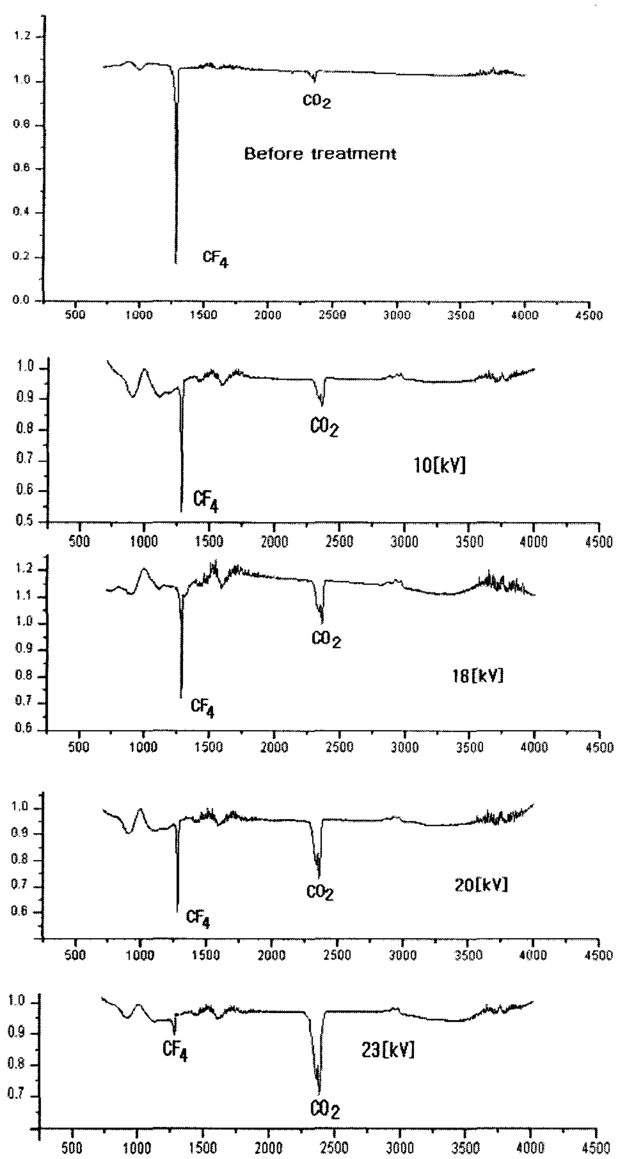

Fig. 10. The FTIR spectra as applied voltage [500 ppm].

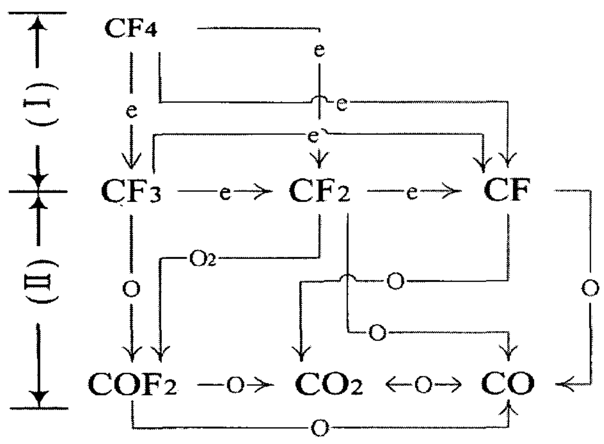

Fig. 11. Major pathway leading to CF_4 conversion.

4. CONCLUSION

In this paper we studied the effect of reactor structure for CF_4 decomposition. To decompose CF_4 we made three kinds of plasma reactors that generate different type of plasma and CF_4 gas decomposition characteristic is investigated in these reactors. As a result, we could get following conclusion.

(1) Plasma that has high-density energy such as arc is better effective than plasma of glow or streamer for CF_4 decomposition.
(2) When arc is generated, non-discharge area occurs in the reactor and decomposition rate of CF_4 decreases.
(3) When arc like discharge with streamer and arc is made in DBH, high CF_4 decomposition rate is took.
(4) Suitable combination of high-density streamer and arc such as DBH has the most effective discharge type for CF_4 decomposition. The CF_4 decomposition efficiency is more than 95% in case of DBH. DBH should be much better than two reactors investigated for CF_4 decomposition.

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