A New Type of Nonthermal Plasma Reactor Utilizing Ferroelectric Pellets

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A new type of nonthermal plasma reactor utilizing ferroelectric pellets is proposed to generate nonthermal plasma efficiently, which is used for simultaneous control of various pollutant gases. Electric charges stored on ferroelectric pellets by corona discharge between a corona tip and a mesh electrode provide partial electrical discharges among ferroelectric pellets. These partial electrical discharges can enhance partial discharges around the surface of ferroelectric pellets. This method utilizes wide reacting area of ferroelectric pellets and partial discharge. Positive and negative dc voltages are applied to the corona tip to generate partial discharges, and corona currents are estimated to investigate charge storage on ferroelectric pellets as function of time and charge relaxation time constants of ferroelectric pellets. As a result, charge relaxation time, dielectric constants of ferroelectric pellets, polarity of applied voltage and applied time influence partial discharges among ferroelectric pellets.

Key words: Nonthermal Plasma, Ferroelectric Pellet, Corona discharge, Charge Relaxation Time Constant, Dielectric Constant.

I. Introduction

ir pollution control can be achieved by both precipitat-A ing dust and pollutant gas control. NOx, SOx, CO2, and VOCs are the main contributors to the environmental problems facing the world, such as acid rain, global warming, ozone depletion, and smog. It has already been demonstrated that nonthermal plasma techniques offer an innovative approach to a cost-effective solution to these problem^{1,3)}. The key to success in nonthermal plasma techniques is to generate a partial discharge in which the majority of the electrical energy goes into the production of high energetic electrons, rather than into gas heating. Energetic electrons rarely collide with pollutants in atmospheric pressure discharges, but mainly with the dominat N₂, O₂ and H₂O, and produce radicals such as O (and O₃) and OH. These radicals have relatively longer lifetimes than electrons and react effectively with contaminant molecules to form nontoxic ones.

Ceramic based on perovskite-type titanates and related materials such as barium titanates and lead-based perovskite are characterized by a broad maximum for the temperature dependence of the dielectric constant and dielectric dispersion in the transition region ⁴⁻⁶. It is reported that partial discharges between ferroelectric barriers take place at very low voltages and catalytic chemical reactions can be expected on the ferroelectric pellet surface.^{7,81}

Ozone is one of the strongest reacting agents and its important property is that it decays without residues that can be harmful to the environment. Also since the on-site production of ozone requires only air and electricity, there is no transport of potentially dangerous chemicals in the application of ozone.

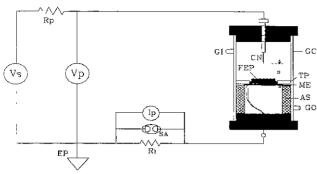
These are the main reasons why ozone is increasingly used for oxidzing purposes as well as being considered for the elimination of NOx from the flue gases of power plants⁹. Thus, ozone generation is used as one of the most important examples of plasma synthesis¹⁰.

In this study, a new type of nonthermal plasma reactor utilizing ferroelectric pellets is proposed to generate nonthermal plasma efficitently, which is used for simultaneous control of various pollutant gases. Electric charges stored on ferroelectric pellets by corona discharge provide partial electrical discharges among ferroelectric pellets. These partial electrical discharges can enhance partial discharges around surface of ferroelectric pellets. This reactor utilizes wide reacting area of ferroelectric pellets and partial discharge between corona tip and mesh electrode.

Thus, the effect of ferroelectric pellets on discharge characteristics and ozone generation of the plasma reactor was investigated based on experimental results. Also, the effects of charge relaxation time, dielectric constants of ferroelectric pellets, polarity of applied voltage and applied time on partial discharges and ozone generations among ferroelectric pellets were investigated.

II. Experimental Apparatus and Methods

Fig. 1 shows schematic diagram of the plasma reactor and exerimental setup. The plasma reactor using ferroelec-



BP: brass plate FEB: ferroelectric-ball : earthed point SA : surge arrestor MS : mica-sheet SB: stainless bolt GC: glass cylinder PR: plasma reactor SP: stainless plate GI: gas inlet TP Ri : current resistor : Teflon plate $(1.00k\Omega)$ GO : gas outlet : protection resistor $V_{\mathbf{p}}$ Rp : pulse power $(750k\Omega)$: pulse current Ιp : gap spacings : pulsed power supply

Fig. 1. Experimental setup.

tric pellets consisted of a corona tip-to-mesh electrode and a barrier of ferroelectric pellets. A Teflon insulating plate (thickness: 2.0 mm) with a hole (diameter: 50 mm) in its center was located on the mesh electrode, and a ferroelectric pellet barrier (FEP, Diameter: 2.5~3.0 mm) was set within the hole of the Teflon plate. Ferroelectric pellets based on perovskite-type titanates, the dielectric constant of which are 6, 35, 60, 100, 650, 2500 and 10000 were used in the experiment. The air gap between the tip and the mesh electrode, s was fixed at 20 mm. This whole structure was then enveloped in a transparent glass cylinder (GC) with a gas inlet (GI) and an outlet (GO). Positive and negative dc voltage sources were applied between the corona tip and the mesh electrode to generate partial discharges. Corona currents were measured to investigate the charge storage on the ferroelectric pellets by a digital storage oscilloscope (Tektronix Model TDS340A) and a high voltage high frequency probe (Tektronix Model P6015A 1000:1). Dielectric constants and relaxation time constants of ferroelectric pellets were estimated by impedance/gain-phase analyzer (HP Model 4194A). Table 1 shows the

The experiment for polarity effect was carried out in the atmosphere of an electromagnetic shielded dark room at a constant feeding air flow rate of 2 l/min, and that for effect of dielectric constants and relaxation time constants was carried out in dry air (bombed air) at a constant feeding flow rate of 2 l/min. The intensity of the generated plasma was estimated by the output ozone concentration measured

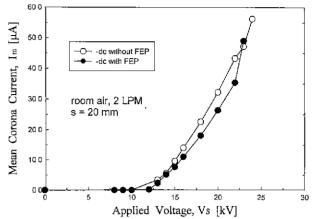


Fig. 2. i, v, characteristics of the plasma reactor.

by an ozone monitor (Dasibi Ozone Monitor, Mode 1108).

III. Results and Discussions

Fig. 2 displays 1_m-v_s characteristics of the plasma reactor with and without a ferroelectric pellet barrier. It shows that only displacement currents flow between the corona tip and the mesh electrode below corona onset voltage. Thereafter, mean corona currents increase with an increasing applied voltage above corona onset voltages. Especially, in the case of a ferroelectric pellet barier, the mean corona current increases drastically after an applied voltage of 20 kV with the generation of intense corona discharges.

Fig. 3 shows the ozone generation of the plasma reactor with and without a ferroelectric pellet barrier as a function of applied voltage. The generated ozone concentration increases linearly with an increasing applied voltage below

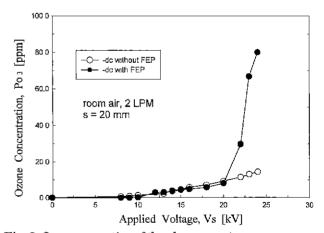


Fig. 3. Ozone generation of the plasma reactor.

Table 1. Capacitance, tan δ, and Time constant of Ferroelectric Pellet for the Experiment

Dielectric Constant [-]	6	35	60	100	650	2500	10000
Measured capacitance [pF]	0.22	0.68	l.71	2.62	16.70	85.88	216.6
$\tan \delta$, D[$\times 10^{-3}$]	40.00	11.80	5.50	3.76	7.36	252.69	11.29
Time constant. $\tau [\times 10^{-6} \text{ sec}]$	36	120	258	378	193	6	126

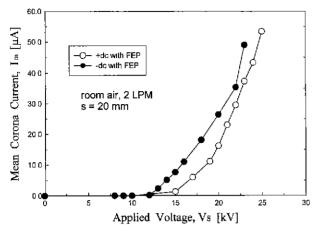


Fig. 4. $i_{\rm m}\text{-}v_{\rm s}$ characteristics of the plasma reactor in the positive and engative corona.

of 20 kV. In the case of the plasma reactor with a ferroelectric pellet barrier, the ozone concentration increases drastically reaching its peak value of 80 ppm with incresing corona currents above the applied voltage of 20 kV.

As shown in Fig. 2 and Fig. 3 the ozone generation increases with the increasing activities of corona discharges on the wide surface of ferroelectric pellets and air gap between the tip-to-mesh electrodes. Thereafter, the generate ozone dissociates due to the heat from the enhanced discharges in the air gap with an increased applied voltage. Thus the plasma reactor with a ferroelectric pellet barrier produces a higher ozone concentration than that without a ferroelectric pellet barrier.

Fig. 4 shows the effects of the polarities of applied voltage on the corona discharge in the reactor with a ferroelectric pellet barrier. Positive and negative corona currents increase gradually with the increase of applied voltages, and increase rapidly after the applied voltages of 15 kV and 12 kV, respectively. However, a negative corona current flows more than a positive one at the same voltage applied.

Fig. 5 depicts the effects of the polarities of applied voltage on the ozone generation in the reactor with a ferroelectric pellet barrier. The generated ozone concentrations in

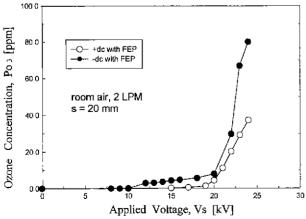


Fig. 5. Ozone generation in the positive and negative corona.

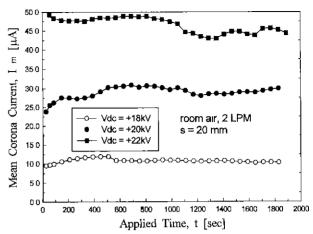


Fig. 6. Variation of the mean corona currents as a function of the applied time.

the positive and negative corona discharge increase linearly below the applied voltage of 20 kV, and increase sharply above the applied voltage due to the enhanced corona discharges in the reactor. Also, ozone in a negative corona discharge is generated more than that in a positive one at the same voltage applied. Fig. 6 shows the variation of the mean corona currents in the plasma reactor with a ferroelectric pellet barrier as a function of the applied time of positive dc voltages. The mean corona current increases with the increase of the applied voltage. The variation of the mean corona current becomes more remarkable as the applied voltage increases as shows in Fig. 6.

This indicates that the storage and discharge of space charges on the ferroelectric pellet barrier repeat in the process of corona discharges.

Fig. 7 displays the variations of the generated ozone concentration in the plasma reactor with a ferroelectric pellet barrier as a function of the applied time of positive dc voltages. The variation of the ozone concentration shows almost the same tendency as that in Fig. 6. However, in the case of negative corona discharges, there is little variation

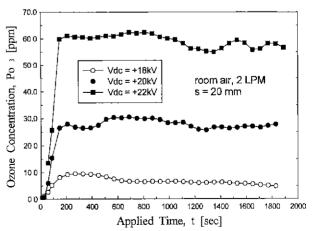
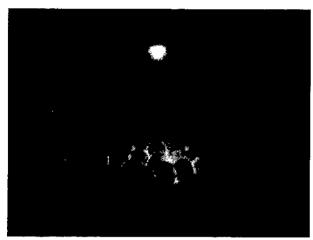
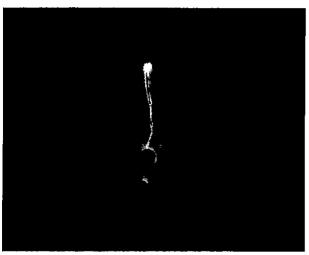


Fig. 7. Variation of the ozone generation as a function of the applied time.



(a) Corona discharge on the ferroelectric pellets



(b) Are discharge on the ferroelectric pellets

Fig. 8. Discharge phenomena in the plasma reactor with a ferroelectric barrier (a, b)

of the mean corona current and the generated ozone concentrations as a function of the applied time of voltages.

Fig. 8 shows discharge phenomena in the plasma reactor with a ferroelectric barrier when a dc voltage is applied. From the observations of the discharge phenomena of a ferroelectric pellet barrier, one can find corona discharges taking place in the upper and lower parts of the ferroelectric pellets when increasing the applied voltage, which produces nonthermal plasma effectively. By incresing the voltage higher, an arc discharge occurs suddenly at the top of the one of ferroelectric pellets from the corona tip. There is hardly any ozone generation here yet the thermal dissociation of the generated ozone can be drastically accelerated.

Fig. 9 shows the effect of the charge relaxation time constant of ferroelectric pellet on the minimum corona sustain voltage, which sustains corona discharge around the tip electrode. The corona sustain voltage increases with increase of the charge relaxation time constant. This indicates that the larger the charge relaxation time constant,

the shows the discharge of the built up charges on the ferroelectric pellets. Thus, the applied voltage must be increased to sustain corona around the tip in case of the pellet with the high charge relaxation time constnat. In addition, the applied voltage should be increased to sustain corona more in case of positive corona than negative one, as shown in Fig. 9.

Fig. 10 depicts the effect of the dielectric constants of ferroelectric pellets on the corona onset voltages on the barrier in positive corona. The corona onset voltage decreases when the dielectric constant increases. This is because the electric field intensity in the air becomes strong in case of the large dielectric constant due to the electrostatic requirement of constant flux density at the boundary of both dielectrics and air.

IV. Conclusions

A new type of nonthermal plasma reactor utilizing ferroelectric pellets is proposed to generate nonthermal plasma efficiently. The mean corona current and ozone generation as function of applied time, polarity of applied voltage, charge relaxation time, and dielectric constant were investiaged. The results are as follows:

- (1) In the case of the discharge reactor with a ferroelectric pellet barrier, the mean corona current increases drastically when a negative dc voltage applied.
- (2) The ozone concentration generated in the reactor with a ferroelectric pellet barrier, increases drastically in the negative corona discharges.
- (3) The variations of the mean corona current and the generated ozone concentration in the reactor with a ferroelectric pellet barrier become more remarkable as the applied voltage increases.
- (4) The minimum corona sustain voltage increases with increase of the charge relaxation time constant. This indicates that the larger the charge relaxation time constant, the slower the discharge process of the built up charges on the ferroelectric pellets.
- (5) The corona onset voltage on the ferroelectric pellets decreases when the dielectric constant increases, in the case of positive corona. This is because the electric field intensity around the ferroelectric pellet becomes strong in the case of the ferroelectric pellets with a large dielectric constant.

References

- NATO Advanced Research Workshop on Non-thermal Plasma Techniques for Pollution Control, Cambridge Univ., England, Sept, 1992.
- J. D. Moon and G. T. Lee, "Discharge and NO Removal Characteristics of a Wire-to-Cylinder Type Nonthermal Plamsma Reactor," Trans. on KIEE., 45(9), 1324-1330 (1996).
- T. Oda, A. Kumada, K. Tanaka, T. Takahashi, and S. Masuda, "Low-Temperature Atmospheric Pressure Dis-

- charge Plasma Processing for Volatile Organic Compounds"; pp. 227-235 in Proceedings of ESA-IEJ Joint Symp. *Electrostatics*, 1994.
- 4. N. Yasuda, H. Ohwa and S. Asano. "Dielectric Properties and Plase Transitions of Ba($\text{Ti}_{1\text{-x}}\text{Sn}_{\text{x}}$)O₃ Solid Solution," Jpn. of Appl. Phys., **35**, 5099-5103 (1996).
- 5. R. Waser, "Charge Transport in Perovskite-Type Titanates: Space Charge Effects in Ceramics and Films," Ferroelectrics. 151, 125-131 (1994).
- 6. D. M. Smyth, "Ionic Transport in Ferroelectrics," Ferroelectrics, 151, 115-124 (1994).
- 7. A. Mizuno, Y. Yamazaki, H. Ito and H. Yoshida, "AC Energized Ferroelectric Pellet Bed Gas Cleaner," *IEEE Trans.*

- on IA, 28(3), 535-540 (1992).
- S. Futamura, A. Zhang and T. Yamamoto, "Control of Byproduct Distributions in Plasma Chemical Processing of Hazardous Air Pollutants": pp. 1961-1967 in Proceedings of 32nd IEEE/IAS Conf., New Orleans, 1997.
- J. D. Moon, "High Efficiency Ozone Generation Using a Pyramidally Embossed Rod-to-Cylinder Electrode and a Pulse Corona Discharge," *Jpa. J. of Electrostatics*, 16(3), 224-229 (1992).
- 10. T. Oda, T. Takahashi and K. Tada, "Decomposition of Dilute Trichloroethylene by Non-thermal Plasma"; pp. 2019-2026 in Proceedings of 32" IEEE/IAS Conf., New Orleans, 1997.