

NOVEL PLASMA CHEMICAL TECHNOLOGIES - PPCP AND SPCP
FOR CONTROL OF GASEOUS POLLUTANTS AND AIR TOXICS

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Abstract

Pulse Corona Induced Plasma Chemical Process (PPCP) and Surface Discharge Induced Plasma Chemical Process (SPCP) represent novel technologies for generating copious active radicals under ordinary temperature and pressure for control of gaseous pollutants (NO_x, SO_x, VOC's) and air toxics (Hg and other heavy metal vapours, trichloroethane, trichloroethylene, freon, toluene) and odors. These are completely dry processes with a simple construction, and their overall cost (initial and running) is one of the lowest among the similar processes. They can be combined with a wet ESP, a bag-filter with absorbent powder coat, catalyzer, active charcoal bed for final removal of reaction products.

Introduction

Pulse Corona Induced Plasma Chemical Process (PPCP) and Surface Discharge Induced Plasma Chemical Process (SPCP) represent novel technologies for generating highly Non-Equilibrium cold plasma in a gas under ordinary temperature and pressure, in which electron temperature is very high and ion/molecular temperature is low (1,2). Since only electrons can generate radicals in collision with neutral molecules to produce plasma chemical reactions, while ions only raise gaseous temperature to cause sparking and arcing, leading to thermal plasma, only electrons but no ions should be accelerated (1st Condition). This can be made either using a very high frequency, or very short pulse voltage. However, a gradual heating of ions and molecules through collision with electrons, finally

to generate transition to thermal plasma, must also be avoided (2nd Condition). This can be made by using a low pressure with a concurrently greater mean-free path and lower collision frequency in the former (high-frequency discharge), or a repetitive interval of pulses for heat-relaxation for the latter (pulse discharge). In most of the industrial applications, including pollution control, the processing has to be made in an ordinary pressure (3rd Condition), which exclude the use of the low-pressure high-frequency plasma and allows the use of the Pulse Approach only. In the Pulse Approach, however, not only the magnitude of the externally applied electric field, but also the length of the streamer coronas has an essential role. Longer the streamer length, the higher is the activity of the radicals generated, and better is the plasma chemical effect achieved (4th Condition). This is because the electrons generated in plasma accumulate on the streamer head to produce an intense electron space-charge field, and the number of the electrons rises exponentially with length, with a concurrent rise in the space-charge field. This space-charge field can be substantially greater in the vicinity of the streamer head than the externally applied pulse field, to generate copious active radicals locally. Higher the field intensity, the greater is the activity of radicals produced. This electron-enhanced high-field spot traverses the electrode gap in the plasma reactor.

PPCP uses Nanosecond Pulse Coronas in a corona electrode system (corona reactor) in combination with a "Nanosecond Pulse High-Voltage Generator" (rise time $T_r=50-100$ ns; half-tail $T_h=200-1000$ ns; peak voltage $V_p=70-250$ kVp; pulse frequency $f_p=50-200$ Hz), to meet all of the above conditions.

SPCP uses a medium-frequency surface discharges generated on a ceramic-made SPCP Element. These ac surface discharges, although seemingly quite different from the pulse coronas, consist of a large number of nanosecond pulse surface discharges having an adequate length (ca. 5 mm) and occurring around the crests of the ac voltage (peak voltage $V_p=4-6$ kVp; frequency $f=8-15$ kHz). The so-called Silent Discharges used in the conventional ozonizers also consists of a large number of nanosecond barriered space discharges, similar to those in SPCP. But the streamer length is restricted and the discharge zone is difficult to cool to allow the use of a high-frequency, as the discharges occur in gas space having a very low heat conductivity (molecular conduction) compared to SPCP of which the discharge zone is a thin film directly attached to the ceramic base. Hence, the use of this particular discharge is not considered.

The great advantages of both PPCP and SPCP are that they are extremely simple in construction, that they are dry systems, and further that they are cheap systems in their overall costs (initial and running costs) (3).

PPCP and SPCP proved to be very effective in the control of gaseous pollutants (NO , SO_2 , VOC's) and air toxics (heavy metal vapours including mercury vapour, trichloromethane, trichloroethylene, freon, toluene, etc.) and odors (4-11). They can also be used in other applications of Cold Plasma, such as Surface Treatment of Plastic Automobile Bumpers (12), Plasma CVD for generation of ultra fine ceramic particles and ceramic filters (13,14), ozone generation (15).

In this paper are presented the technologies of PPCP and SPCP for gaseous pollution control, together with their test data.

Pulse Coron Induced Plasma Chemical Process (PPCP)

Figure 1 shows a Nanosecond Pulse High-Voltage Generator used for PPCP, consisting of a Control Unit, Power Unit and a Pulser Unit. The Control Unit comprises an inverter circuit for generating a pulse voltage to be fed to the primary of a High-Voltage Transformer of the Power Unit. Its output voltage charges the Pulse-Forming Condensator Bank, C_p , during the period when the Rotary Spark Gap, RSG, is off, both C_p and RSG being contained in the Pulser Unit. RSG connects C_p directly to the Corona Reactor CR when the charging voltage from PS is off. This timing control is made by a photo-sensor/glass-fiber link, fed back to the Control Unit (Figure 2). Figure 3 (a) shows the pulse wave form appearing at the Corona Reactor, but its leading edges are superposed with the ringing voltage as shown in Figure 3 (b) resulted by an LC-oscillation in the load circuit. This oscillation produces voltage enhancement to give an effective pulse peak voltage. The performance data are given in Figure 2.

There are several points to be considered in the application of PPCP. The first one is the nature of the radicals, either oxidizing radicals (O , O_3 , O_2^- , OH) or reducing radicals (H , NH , NH_2 , N , N_2^+), if they induce chain-reactions, and if the radicals are active enough to decompose the pollutant molecules encountered. The second one is the generation rate of the radicals per unit time in the Reaction Chamber in comparison with the pollutants input per unit time. Of course, this ratio can be less than a stoichiometric in the case the chain-reactions can be expected. Whereas it can be greater in the case the radicals used have a substantially shorter life time than the mixing time with pollutant components in gas. The generation rate of radicals is proportional to the pulse frequency, f_p , and the input pulse power, P . The third factor is the gas residence time, T_{res} , in reaction chamber. The decomposition rate rises with T_{res} up to a certain point, beyond which it becomes saturated. This is resulted by the reverse reaction: the decomposition the reaction products back to the original one, possibly by dissociation due to electron collision. This reverse reaction is one of the essential features of plasma chemical reaction, which, however, can be avoided by the co-existence of scavenger components catching or fixing the reaction products (NH_3 for DeNOx and DeSOx; dust particles for DeSOx, etc.). Thus, the scavenger of the reaction products, if it exists in the main gas (e.g. dust), or if it must be added (NH_3), is fourth point. In relation to this factor, the final processing of the reaction products, either in gas form or solid particulate, is the fifth point to be considered: NH_3 -addition plus ESP or bag-filter; bag-filter with dry solvent coat ($Ca(OH)_2$; CaO_2 etc.); wet-ESP with solvent irrigation; catalyzer). All these factors are to be taken into account to achieve the best cost/effectiveness of PPCP.

A careful economical assessment on PPCP to be applied for DeNOx and DeSOx in coal-burning utility boilers, as compared with the conventional ammonia chatalytic process (for DeNOx) combined with the lime scrubbing process (for DeSOx), and an electron beam process for the simultaneous DeNOx and DeSOx, revealed the results estimation being

the minimum for PPCP in its overall cost (installation cost + running cost) (3).

Figure 4 shows the results of laboratory DeNOx test, plotted against the average peak field intensity at 5 cm electrode gap. A gradual drop in NO concentration starts with the corona initiation at $E_p=4$ kV/cm, with a concurrent rise in NO₂ concentration. The latter indicates a peak at $E_p=11$ kV/cm, succeeded by a sharp drop to become 0 at $E=12$ kV/cm (4). Figure 5 shows the DeNOx results with the different polarity, indicating the drop in NO₂ concentration does not occur without co-existence of water molecules, namely the role of OH-radicals which are produced only at a higher field intensity than O-radicals.

Figure 6 shows the effect of the pulse frequency, f_p , on the removal rate of mercury vapour by conversion into HgO solid particulates (5). A definite proportionality can be seen to exist between f_p and the logarithm of the removal rate. Figure 7 indicates for NO, SO₂ and mercury vapour the removal rate plotted against gas residence time, T_{res} , which clearly indicates the saturation tendency as described previously (5). Figure 8 shows for these pollutants the effect of the energy input per unit volume of gas on the removal rate.

The effects of these parameters, P , T_{res} , and f_p can be summarized by the equation:

$$\text{Removal Efficiency} = A(1 - \exp(-k \cdot P \cdot f_p \cdot T_{res})) \quad (1)$$

Figure 9 shows the removal efficiency measured for NO under different conditions of P , T_{res} and gas flow rate, Q , plotted against a parameter $(P/Q)(T_{res})^{-1/2}$ ($=E/T_{res}^{1/2}$) ($\text{kWh}/\text{Nm}^3 \cdot \text{s}^{1/2}$) (4).

It should be noted that the design guide for PPCP system (power supply and reactor) is to produce adequate quantity of radicals to achieve the desired level of the removal efficiency for the given species of gaseous pollutants at their mass-inputs (concentration x gas flow rate) for given operating conditions (temperature, pressure, humidity, etc.).

Surface Discharge Induced Plasma Chemical Process

Figures 10 and 11 indicate the photographs of planar and cylindrical SPCP Elements, respectively, and Figures 12 and 13 their cross-sections. Linear discharge electrodes (tungsten) are attached on the surface of a high-purity alumina ceramic layer, and a planar induction electrode (tungsten) is embedded in its inside, facing to the discharge electrodes. The thickness of the intermediate alumina layer is small (0.5 mm). A medium frequency, medium-high voltage (8-15 kHz; 4-6 kV peak) is applied between the two electrodes to generate the ac surface discharge around its crest in each half-cycle (Figures 14 and 15). As described previously, these ac surface discharges consist of a large number of nanosecond surface streamer coronas. The role of the planar induction electrode is to enhance the length of these streamers by providing the streamer tips with the tangential field of adequate intensity at any instant during its development. Thus, an adequately long streamers can be generated along the alumina surface with an essentially lower voltage. In addition, the initial electron triggering streamers is that trapped in the surface state (trap) on the alumina plate, and is freed only at an

instant when a high local field intensity is reached. Furthermore, with the existence of alumina dielectric, a field intensification occurs in the gas space. As a result of these phenomena, radicals having a higher activities are generated in this particular mode of discharge.

Two different modes of application exist in SPCP-control of gaseous pollutants. One is the Direct Mode Treatment, where the gas to be cleaned passes through the radical-active region in or near the surface plasma to subject to radical reaction. Another one is the Indirect Mode Treatment, where a suitable gas is passed along the plasma region to generate desired species of radicals, and these radicals are fed into the main stream of polluted gas for radical reaction. In the latter mode, a high gas speed with a concurrently short contact time is generally required to provide the best decomposition of pollutants, as the active radicals mostly have very short life time, with the exception of nitrogen radicals (several tens seconds). Care must be taken in the application and design of SPCP reactor: species of carrier gas and pollutants; concentration of the pollutants; radicals to be generated; reaction products and their final removal; temperature and pressure; direct-mode or indirect-mode, etc.). For instance, generation of excessively active oxidizing radicals not only turn NO into NO₂ and further product of higher oxidation, but also generate NO by oxidation of N₂ in gas. Similarly, excessively active nitrogen radicals, a kind of reducing radical, generate NO from O₂ in gas.

Figure 16 shows a standard SPCP-Reactor with a water jacket for cooling, and Figure 17 a standard power supply with a fixed output voltage but variable frequency, possible to drive 6 pieces of the SPCP-reactors (Figure 16) in parallel. The SPCP-reactor represents a kind of non-linear load with its inter-electrode capacity indicating a jump in each half-cycle when surface discharge occurred. As a result of this non-linear behaviour, current indicates a sudden great increase when a certain critical voltage is exceeded (jumping phenomenon), to burn the transformer winding. This particular power supply has a special circuit design to avoid this non-linearity induced burnout.

Figures 18 to 21 indicates the DeNOx performance by SPCP measured under various conditions (6), which indicate basic features of SPCP gaseous pollution control. A very high DeNOx performance can be easily achieved even for a high inlet concentration when the carrier gas has no oxygen (Figure 18). With the increase in oxygen from 3 % to 11 % the DeNOx rate for ca. 1000 ppm NO carried by N₂ drops from 100 % down to 60 %. (Figure 19), which suggests the importance of excess air in combustion gas. Furthermore, adequately high DeNOx performance can be obtained even with a very short gas residence time $T_{res}=0.002$ s (Figure 20). Also, the Indirect-Mode Operation using SPCP-activated air indicates almost the same DeNOx performance as that of Direct-Mode Operation.

Figure 22 indicates a photography of the SPCP Deodorizer, and Figure 23 its construction, where a Venturi-Type Reactor is used so that the entire gas passes through the radical active region in its throat with a very short contact time (11). A very satisfactory deodorizing performance is achieved in professional areas with strong odors, including incineration plants of municipal wastes.

Figures 24 and 25 shows the results of freon decomposition tests (CFC-113) (8) continued after the previous tests (7). Figure 24 shows a substantial performance increase obtained by increasing frequency to 50 kHz from the previous 5 kHz, and the effect of the discharge induced heating of the SPCP-Element measured with its cooling fan ON and OFF. It can be seen that the heating provides a great positive effect for SPCP freon decomposition in contrary to SPCP Ozone Generation (15, 16). The advantage of using a higher frequency lies in its increase in energy consumption, as shown in Figure 25. It ranges for 50 kHz from 1.5×10^9 (J/mol) at $T_{res}=0.5$ s to 9.5×10^9 (J/mol) at $T_{res}=4.5$ s.

Figure 26 indicates for trichloroethylene with 333 ppm concentration the rate of decomposition plotted against the gas residence time, T_{res} (9). Figure 27 indicates the similar plots for acetone with 1000 ppm concentration, and Figure 28 those for 2-propanol (9).

Figure 29 shows for toluen the rate of decomposition at $T_{res}=9$ s plotted against the power input (10). In this case a time-dependent accumulation of tar-like reaction product is observed, and it stopped the surface discharge after 1 hour operation in the case the toluen concentration is as high as 2000 ppm. However, this tar-like film is decomposed by passing carrier gas only (dry air), to disappear after 6 hours.

Thus, it is concluded that SPCP can effectively decompose various hazardous and toxic gaseous pollutants emitted from working places.

Conclusion

The plasmachemical backgrounds for PPCP and SPCP for control of gaseous pollutants and air toxics, with their test results. These technologies provides cost-effective control means of dry processing. PPCP is suitable for a medium to large gas volume where the cost of Nanosecond Pulse High-Voltage Generator can be offset by a large electrode gap possible in the Corona Reactor. SPCP is suitable for a small to medium gas volume where the lower initial costs of both the SPCP-Reactor and its power supply are the most essential factor. The first practical application of PPCP was the surface treatment of polypropylene automobile bumper, from which adequate knowledge and experience could be obtained for pollution control in both Corona Reactor and Pulse Power Supply. A series of tests are being performed at a waste incineration plant of Osaka for PPCP control of dioxine, and an encouraging results are being obtained. The first practical use of SPCP was ozone generation, followed by a SPCP deodorizer, both being very cost-effective. Another series of tests for SPCP control of hazardous and toxics gases are being continued.

Acknowledgements

The authors are very grateful to the warm support and useful advises given to the PPCP research from JAMDA, and Mrs. Sakakibara, Kito and Saiki of Takuma Co., Ltd. Thanks are also due to Dr. Takahashi, Mrs. Tochizawa, Kuwano and Akutsu of Nippon Paint Co.,

Prof. Oda of University of Tokyo, Mr. Tabata and Mr. Yamaguma of the Institute of Industrial Safety, Ministry of Labor Japan for their supports and advises given to the SPCP research.

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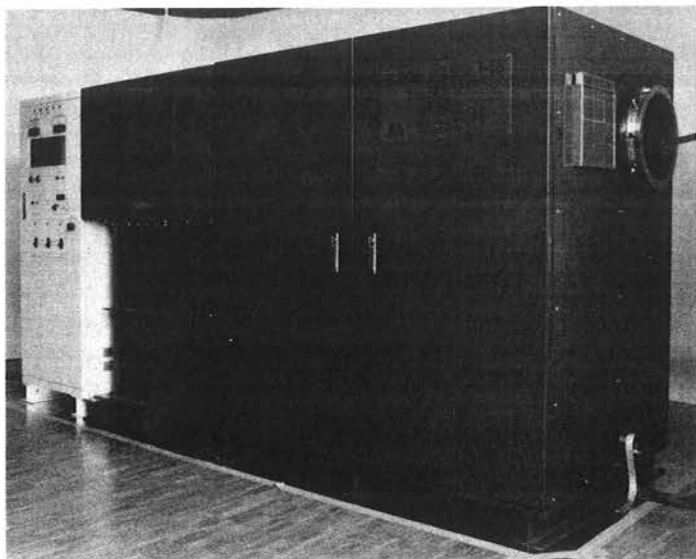
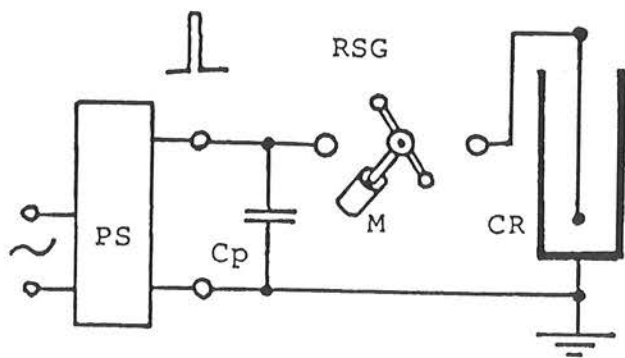


Figure 1

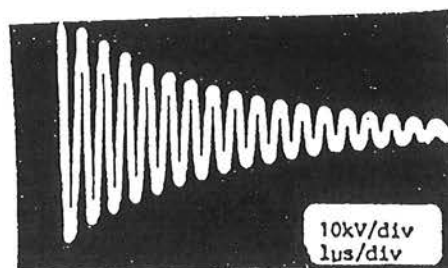
Nanosecond Pulse High-Voltage Generator for Testing (ELDYNE PULSER T-5S)
 capacity: $S=5m^2$ (total area of Corona Reactor)
 rise time: $T_r=100ns$
 half-tail: $T_h=500ns$
 peak voltage: $V_p=100kV$
 frequency: $f_p=20-200Hz$



PS : Charging Pulser
 Cp : Pulse Forming Condenser
 RSG : Rotary Spark Switch
 M : Servo-Motor
 CR : Corona Reactor



(a) Saw-teeth pulse waveform



(b) Oscillation part

Figure 2 Circuit Diagram of Pulser

Figure 3 Pulse Wave Form

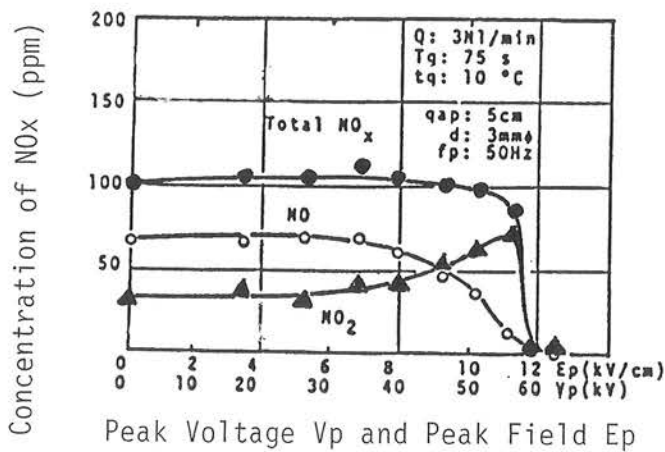


Figure 4 Effects of Peak Voltage and Peak Field on DeNOx Performance (NH₃(180ppm) added; dry air; negative)

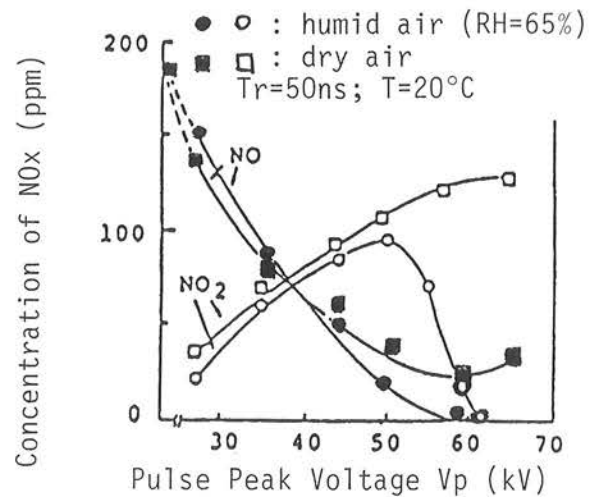


Figure 5 Effect of Moisture on DeNOx Performance (positive polarity)

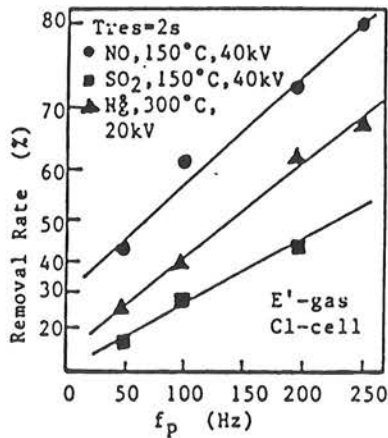


Figure 6 Effect of Frequency

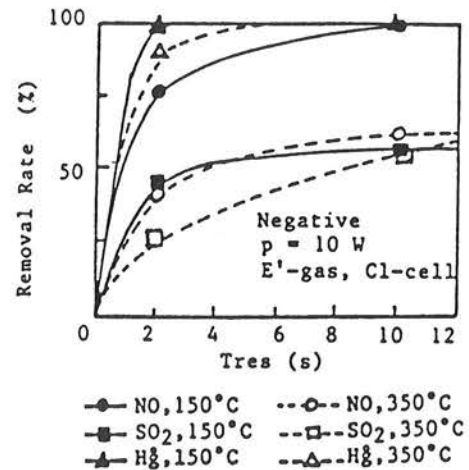


Figure 7 Effect of Residence Time

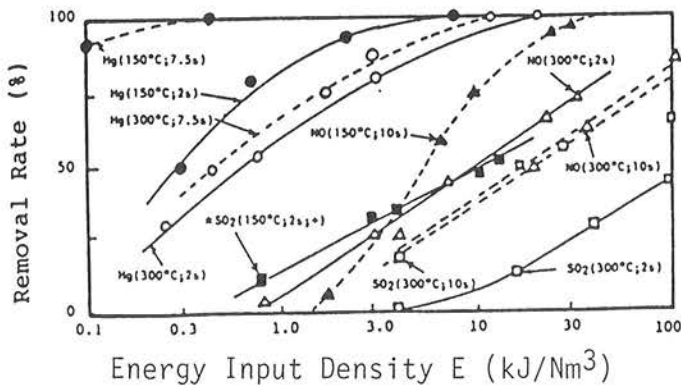


Figure 8 Effect of Energy Input Density on DeNOx, DeSOx and Mercury Removal Rate

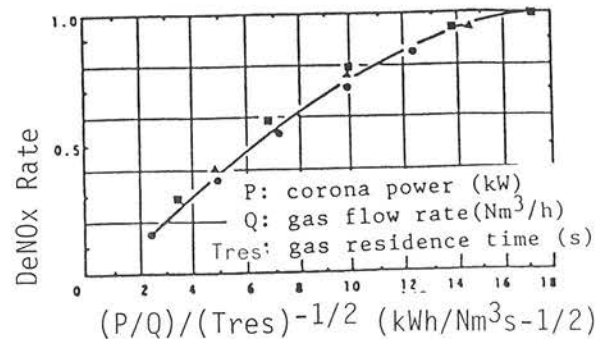


Figure 9 DeNOx Rate vs. Performance Parameter $(P/Q)(Tres)^{-1/2}$

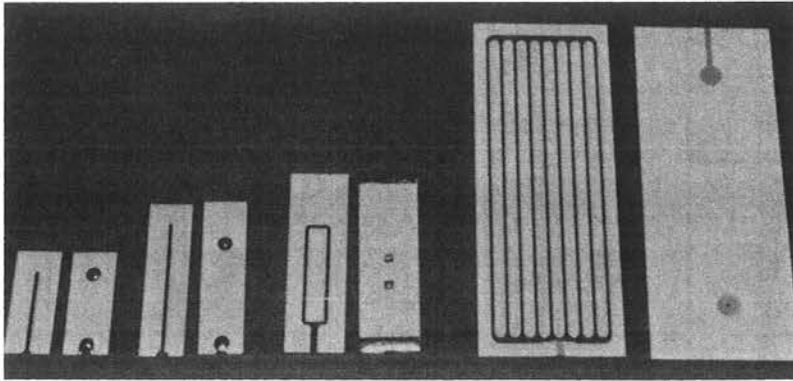


Figure 10
Planar Type SPCP Element

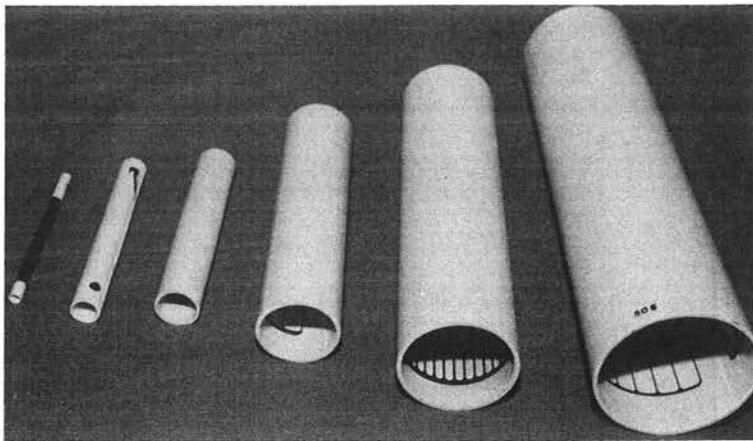


Figure 11
Cylinder Type SPCP Element

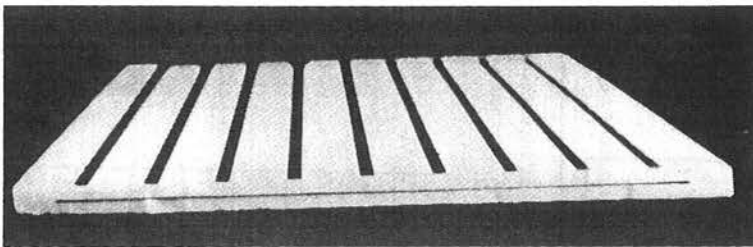


Figure 12
Cross-Section of Planar
Type SPCP Element

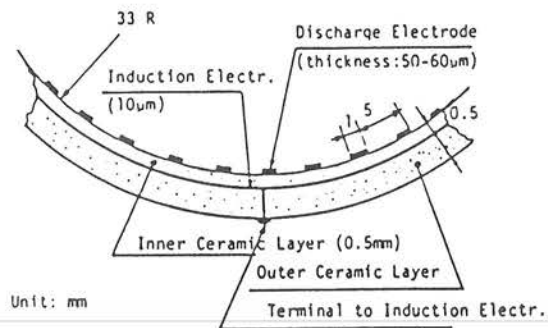


Figure 13
Cross-Section of Cylinder
Type SPCP Element
(OC-70-A)

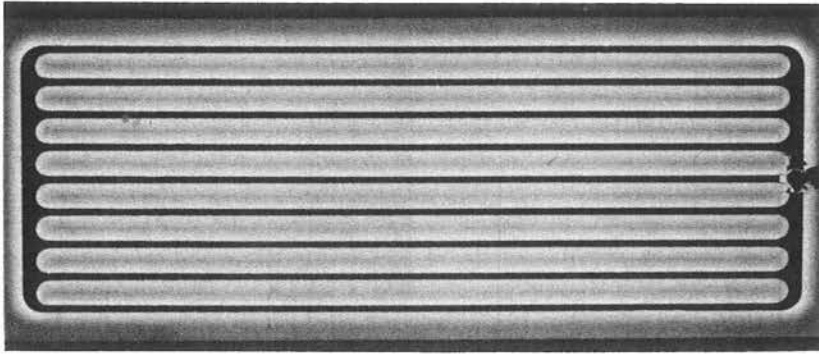


Figure 14
Medium-Frequency Surface Discharge on A
Planar SPCP Element

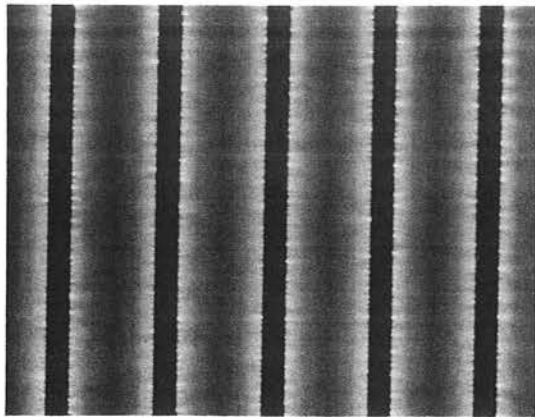


Figure 15 Surface Streamers

Figure 16 SPCP-Reactor with Water Jacket for Cooling →

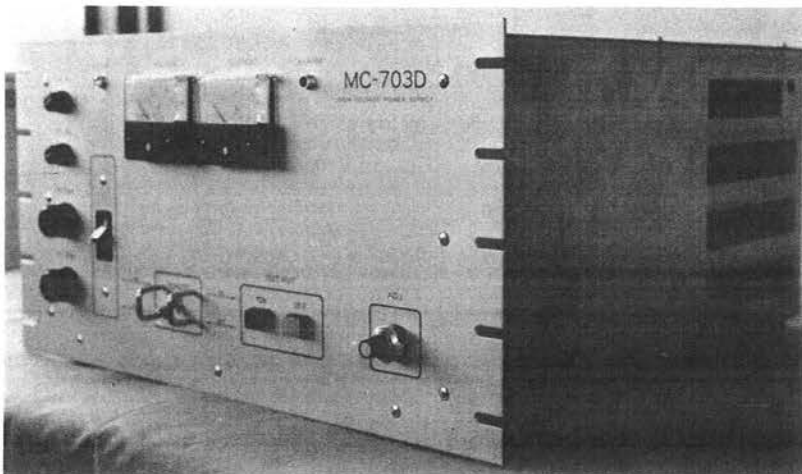
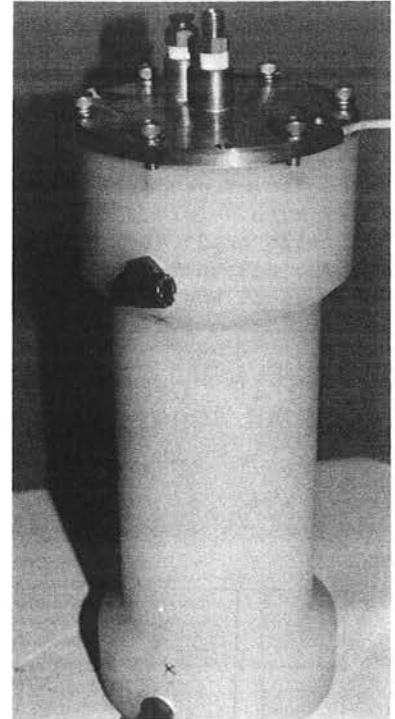


Figure 17
Standard Power Supply
for Driving 6 SPCP-
Reactor in Fig. 16 with
Fixed Voltage and Vari-
able Frequency (MC-703D)

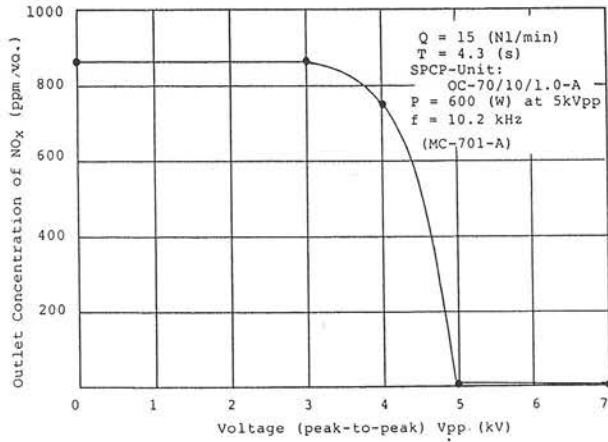


Figure 18 DeNOx Performance for Nitrogen Carrier Gas in SPCP at Longer Tres

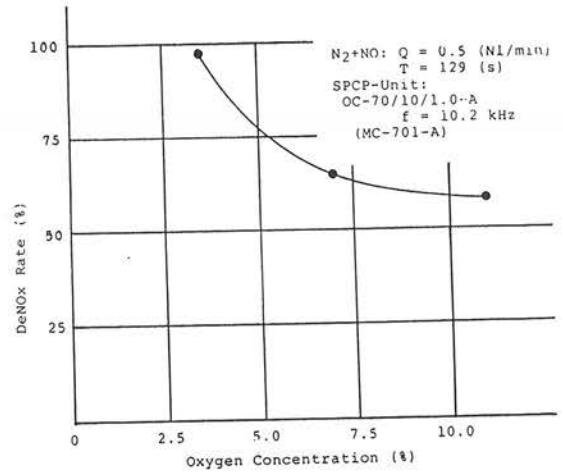


Figure 19 Effect of Oxygen in Carrier Gas on DeNOx Performance

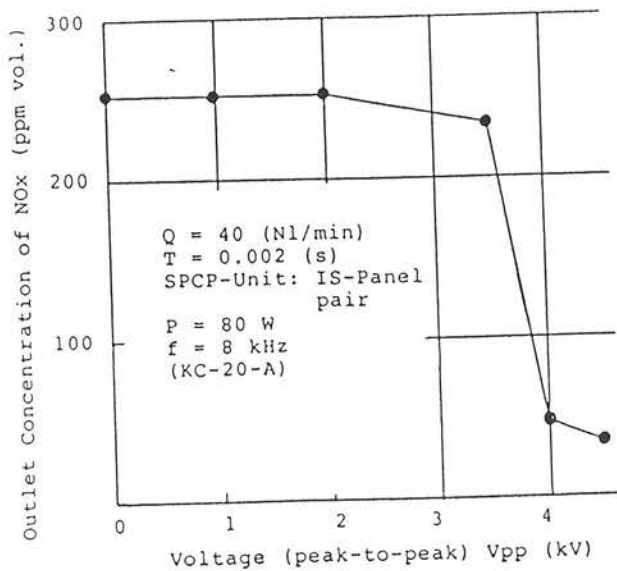


Figure 20 DeNOx Performance of SPCP for Air/N₂/NO System at Tres=0.002 s

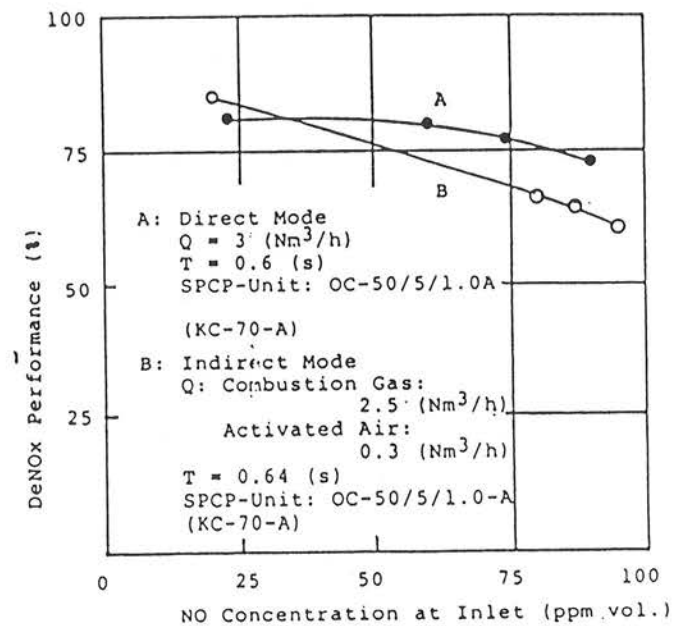


Figure 21 DeNOx Performance of SPCP vs. Inlet NO Concentration for Direct and Indirect Mode Operations (oil-burning)

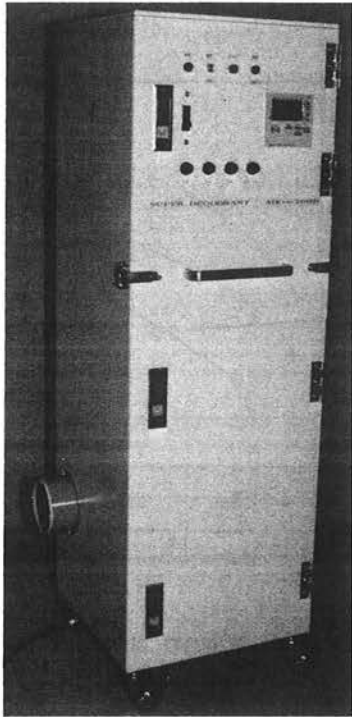
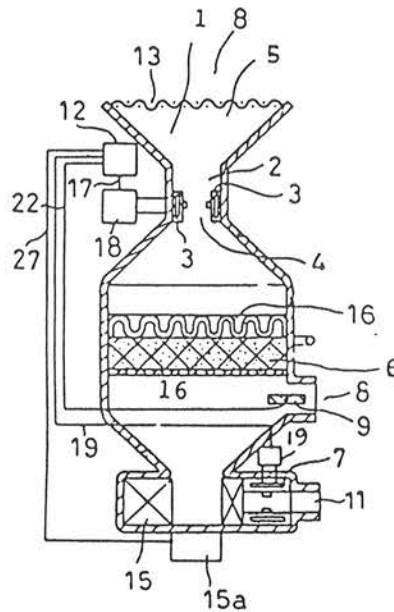


Figure 22 SPCP Deodorizer (ADO-250-A)



- 1: Venturi
- 2: Throat Part
- 3: SPCP Elements
- 4: Radical-Active Regio
- 5: Air Inlet
- 6: Catalyzer
- 7: SPCP Elements for Ozone Fumigation
- 8: Air Intake for Fumigation-Mode Operation
- 9: Flapper
- 11: Clean Air Outlet
- 12: Timer
- 13: Prefilter
- 15: Suction Fan
- 15a: Fan Motor
- 16: Mini-Pleats Filter
- 17-27: Lead Wires
- 18: HF-HV Power Supply
- 19: HF-HV Power Supply

Figure 23 Construction of SPCP Deodorizer

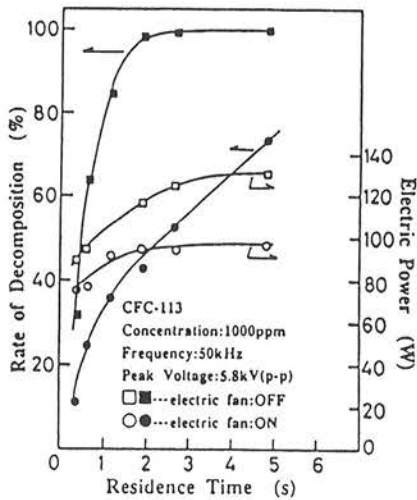


Figure 24 SPCP Decomposition Performance of CFC-113 at 50 kHz with Cooling Fan ON and OFF

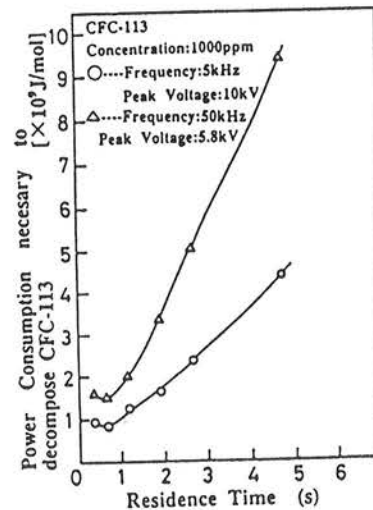


Figure 25 Energy Consumption for SPCP Decomposition of CFC-113

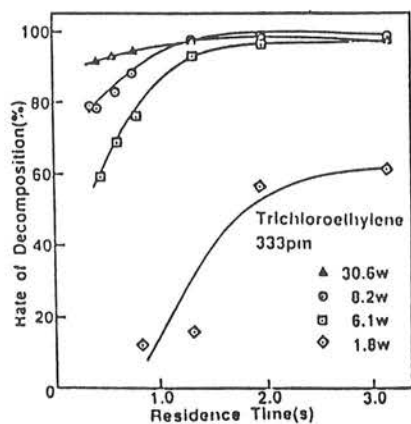


Figure 26 Decomposition Performance of SPCP for Trichloroethylene (333 ppm)

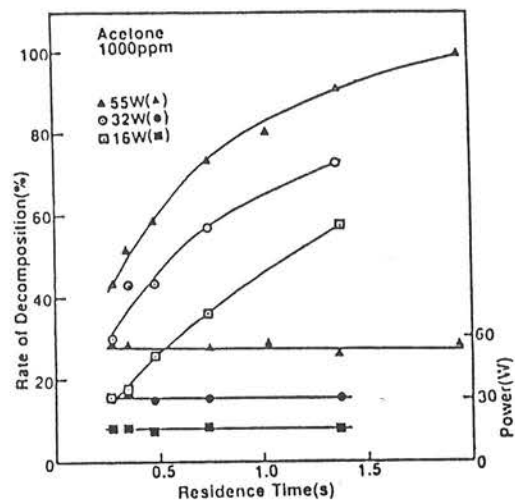


Figure 27 Decomposition Performance of SPCP for Acetone (1000 ppm)

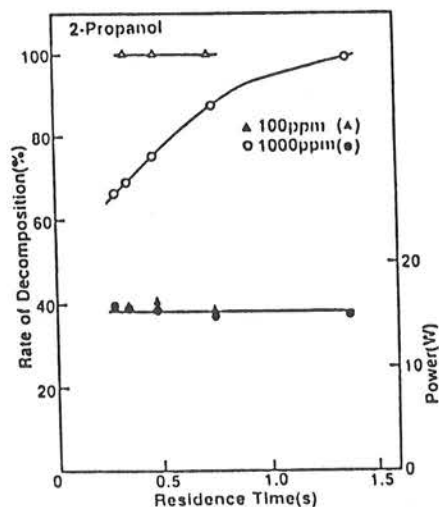


Figure 28 Decomposition Performance of SPCP for Isopropylalcohol (100 ppm; 1000 ppm)

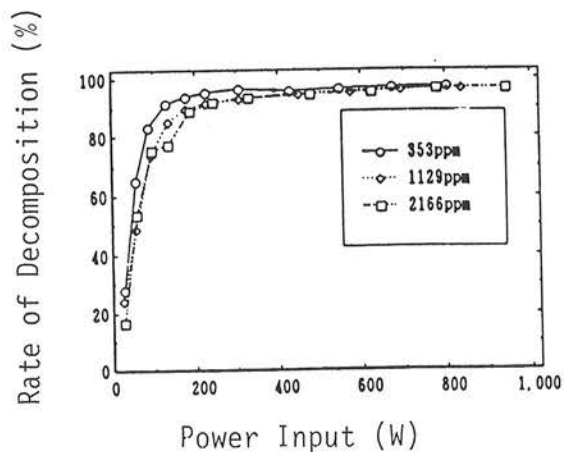


Figure 29 Decomposition Rate of Toluene vs. Power Input by SPCP (Tres=9 s)