

Development of 0.5 MWe Scale DeSO_x-DeNO_x System Using Pulsed Corona Discharge

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ABSTRACT

To verify the feasibility of a simultaneous removal process of SO₂ and NO_x in combustion flue gases by pulsed streamer coronas, an industrial-scale pilot plant that can treat 1,800 Nm³/h of coal-fired combustion flue gas has been developed and tested. The system consists of a nonthermal plasma reactor that is similar to an existing electrostatic precipitator with wire-to-plate configuration, and a pulse generator that can generate output pulses of 130 kV, 500 ns, and 100 J per pulse. A MPC technique using Metglas magnetic switches was adopted to meet an acceptable level of lifetime and reliability for practical use of the process. The total energy consumption in this process is slightly higher than that of conventional methods. Therefore, as a part of energy saving, this paper deals with impedance matching conditions between the pulse generator and the plasma reactor, and reports some experimental results for removal of SO₂ and NO_x with and without C₂H₄ as a additive. The addition of C₂H₄ significantly enhanced the removal performance of NO_x. The impedance characteristics of the pulse generator are also analyzed by experiments.

I. Introduction

The nitrogen oxides (NO_x) and sulfur dioxide (SO₂) in flue gases exhausted from coal-burning power plants, iron and steel plants, paper mills, internal-combustion engines are the major air pollutants. These pollutants are the cause of acid rain and photochemical smog and are harmful to human beings, so some countries have restricted their emissions.

Many control devices are available, but due to increasingly stringent regulations, conventional devices tend to be expensive and huge. Therefore, a large market will be opened up because of the requirements for new regulations.

Nonthermal - low temperature or nonequilibrium - plasma techniques among others have attracted our attention since 1970'. They can be obtained by producing a plasma in which a majority of the electrical energy goes into the production of energetic electrons. The electron mean energies are, therefore, considerably higher than those of the ions and the molecules.

There are many types of nonthermal plasma that have been investigated and developed for environmental applications. But it can be created in essentially two different ways, electron-beam irradiation and electrical corona discharge [1]. Although work on the electron-beam process started a decade earlier than that on the corona discharge process, the corona method has the advantages of being modular, so it can be

used on a large or a small scale and can much more easily be retrofitted in existing power plants. In addition, it has a lower capital cost and no x-ray radiation shielding. Recently, it is recognized that the nonthermal plasma technique, especially a pulsed streamer corona method in applications of removal of SO₂ and NO_x has several features such as being dry, good use of end products, simultaneous treatment, easy processing control, and no stack reheat. It is particularly efficient only when the pollutant is present in very small concentrations.

Electrical corona discharge methods can be implemented in many ways, depending on the geometry of the plasma reactor and the electrical power supply (DC, AC, or pulsed). The types of electrical discharge reactors for the treatment of contaminated air include pulsed corona, ferroelectric bed, dielectric-barrier discharge, and surface discharge. Since pulsed corona reactors with a wire-to-plate configuration have shown very promising results and could be retrofitted to an existing electrostatic precipitator, it would be especially attractive.

Generally, streamer corona discharge energized by a pulse voltage generator with a fast rise time can produce an intensive plasma, which effectively promotes gas-phase chemical reactions. Therefore, uniform streamer formation is necessary to achieve high performance in the streamer corona treatment. In the case of negative polarity, the ionization zone is limited to the vicinity of the discharge electrode and streamers do not propagate across the electrode spacing. However, positive streamers can propagate across the electrode spacing, ionizing the treatment volume to a larger degree and more uniformly [2]. Previous test results [3] show that the SO₂ removal efficiency is higher than that of negative corona when applied.

Until now, both laboratory and pilot scale tests have been conducted in some countries [2~7]. Those results have indicated significant technical and economical advantages of the nonthermal plasma process based on the pulsed corona discharges compared to the electron beam process and other conventional methods. In 1988, industrial-scale experiments on the use of the pulsed streamer corona process for the simultaneous removal of NO_x and SO₂ from combustion flue gas have been carried out by the Italian National Electricity Board (ENEL) at the coal-burning power station in Marghera, Italy [4]. According to the results, the energy consumption rate was up to 5 % of electric power generated by the power plant. The energy consumption required by the process represents the largest operating cost and the transfer efficiency of the electrical energy from a wall-plug to the plasma in a reactor represents a key factor for the economical competitiveness of the process.

It is indispensable to enhance the chemical reaction efficiency and to reduce the energy consumption since the total energy consumption in the process depends both on the chemical efficiency of the plasma and the energy conversion efficiency from the main power source to the corona streamers. From the viewpoint of energy saving, for example, the injection of chemical additives should be considered and the energy efficiency in a pulse generator also could be increased up to 80 %.

II. Experimental Setup

1. Pulsed streamer corona reactor

The corona reactor designed and manufactured in this work is very similar to an existing EP (Electrostatic Precipitator) with wire-to-plate geometry as shown in Fig. 1. It is comprised of two fields, each field contains four channels with a plate distance of 160 mm, a length of 1.6 m, and a height of 1.5 m. The discharge part of the reactor consists of a rectangular frame with 9 wire in each diameter of 3 mm, which is placed vertically at the center line between the plates.

The capacitance of the reactor in connection with the inherent geometry was estimated in the vicinity of 3 nF. With this value and a rise time of output pulse voltage generated from the pulse generator, the maximum permissible inductance in the discharge circuit can be calculated as follows [5]:

$$L = \frac{(3 \cdot t_r)^2}{P^2 \cdot C} = \frac{(3 \times 100 \times 10^{-9})^2}{P^2 \times 3 \times 10^{-9}} \approx 3 \text{ mH} \quad (1)$$

where, t_r : the rise time of pulse voltage
 C : the capacitance of the reactor.

Satisfying that value and reducing the stray inductance, it is necessary to construct a very compact reactor and to connect a generator to a reactor as short as possible. The latter means that a connection lead cable should be straight and short. Detailed characteristics of the reactor are summarized in table 1.

Table 1. Main characteristics of the pulsed corona reactor.

Gas flow rate	1,800 Nm ³ /h, max.
Emitting wire diameter	3 mm, round type
Distance between plates	160 mm
Distance between wires	130 mm
Number of channels	4
Number of fields	2
Total length of wires	108 m

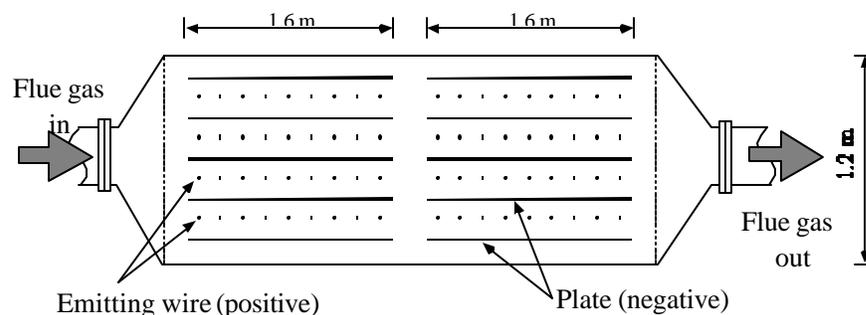


Fig 1. HANJUNG pulsed streamer corona reactor.

2. High power pulse generator

Taking account of the previous test results [4, 8], the optimal condition for a pulse generator requires that it should be able to generate a pulse voltage of 150 kV peak, pulse current up to 10 kA peak, a pulse width of 500 ns at FWHM (full width at half maximum), and approximately 800 J per pulse. It must, moreover, run continuously at pulse repetition rates between 100 pps (pulse per second) and 300 pps, and have at least a maintenance interval of 6 months as a baseline. The lifetime and reliability of the system are extremely important as downtime for maintenance affects plant availability, either scheduled or unscheduled.

It is obvious that one of the key components for a pulse generator to meet those specifications is the switch system. Some of switches are available commercially for pulse power applications. However, a magnetic switch that has solid-state and nonlinear inductance characteristics is the only one to achieve completely the above specifications because other switches, in general, have poor lifetimes and higher cost levels [5,9]. Based on Melville pulse compression networks [10], the MPC (Magnetic pulse compression) method using saturable reactors and pulse forming capacitors is a useful technique to make high power pulses having a narrow width and a steep rise front. The main features of the MPC type pulse generator are a long lifetime and high reliability because of its solid-state properties. These are very important to the industrial applications.

Fig. 2 shows a simplified circuit of the pulse generator developed with a high voltage DC power supply (Maxwell model CCDS850P1480, 4 units paralleled), MPC switches (Metglas 2605CO), and a pulse transformer. The use of a pulse transformer and a constant current capacitor charging type HVDC power supply makes the generator simple, reliable, and efficient. Especially a DC power supply adopting the above type can eliminate much of the energy loss assessed in conventional resistive charging of an initial stage capacitor with a constant voltage power supply.

The operation principle of Fig. 2 is as follows. First, the capacitor C_0 is charged by the HVDC power supply up to 50 kVmax. Charged energy in C_0 is transferred to C_1 through L_0 , the charging inductor, by switching of SW_0 (thyatron, EEV model CX1525A) during 3 μ s. After charging C_1 , MPC1 is saturated and the energy stored in C_1 is transferred to C_2 through the pulse transformer. The output pulse from C_2 is compressed again by MPC2 (500 ns, F

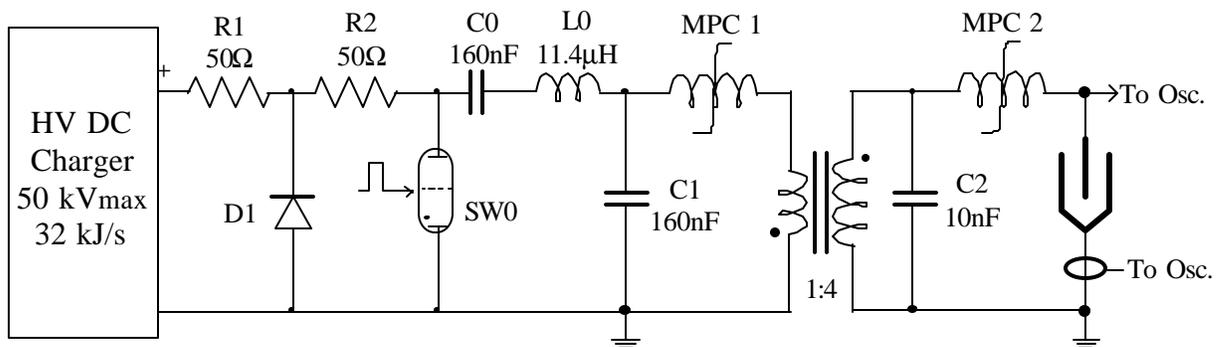


Fig 2. Schematic circuit diagram of the MPC pulse generator.

Table 2. Design parameters of the MPC pulse generator.

Maximum output power	30 kW
Peak output voltage	100 ~ 130 kV
Peak output current	5 kA
Pulse energy	100 J
Maximum repetition rate	300 pps
Pulse width (FWHM)	500 ns
Pulse rise time (10 ~ 90 %)	100 ns
Efficiency	~ 80 %
Maintenance interval	$> 3 \times 10^9$ pulses

WHM). In order to protect the HVDC power supply against voltage reversal, resistor R_1 and R_2 and diode D_1 are inserted. The generator can deliver about 100 J per pulse with pulse repetition rate up to 300 pps to the plasma reactor. Table 2 summarizes the design parameters of the pulse generator.

3. Pilot plant and measurement systems

The layout of the pilot plant built in HANJUNG is shown in Fig. 3. These facilities have the gas cleaning capacity of 1,800 Nm³/h, equal to gas capacity of 0.5 MW, which is bypassed from the main stream of the pulverized coal combustion system with 2,500 Nm³/h. This facility is composed of a bag filter to collect fly ashes from the flue gas, a heat exchanger to control temperature at the inlet of the reactor, the pulsed streamer corona reactor energized by the MPC pulse generator, an EP to collect ammonium salts produced by the process, and an induced draft fan.

In this work, additional SO₂ and NO gases are injected into the flue gas at the downstream of the heat exchanger to simulate the real coal-fired combustion flue gas compositions, and then ammonia and a chemical additive, C₂H₄, are injected into the upstream of the reactor.

The output pulse voltage and current are measured using a digital oscilloscope (Tektronix model TDS744A) having an analog bandwidth of 500 MHz and a sampling rate of 2 GS/s, a 4000:1 resistive voltage divider (Haefely Trench model 550483), and a wideband current transformer (Pearson model 4997). All the signal waveforms acquired by the oscilloscope are saved for analysis on such things as electrical diagnostics and energy estimations.

Power input to the HVDC power supply was also monitored using a three-phase watt-hour meter so that a wall-plug efficiency of the reactor could be calculated. Gas analysis with regard to SO₂ and NO_x (= NO + NO₂) was performed with a chemiluminescent NO_x analyzer and a pulsed UV fluorescent SO₂ analyzer (both Thermo Environmental Co.). The temperature was measured at both inlet and outlet of the reactor.

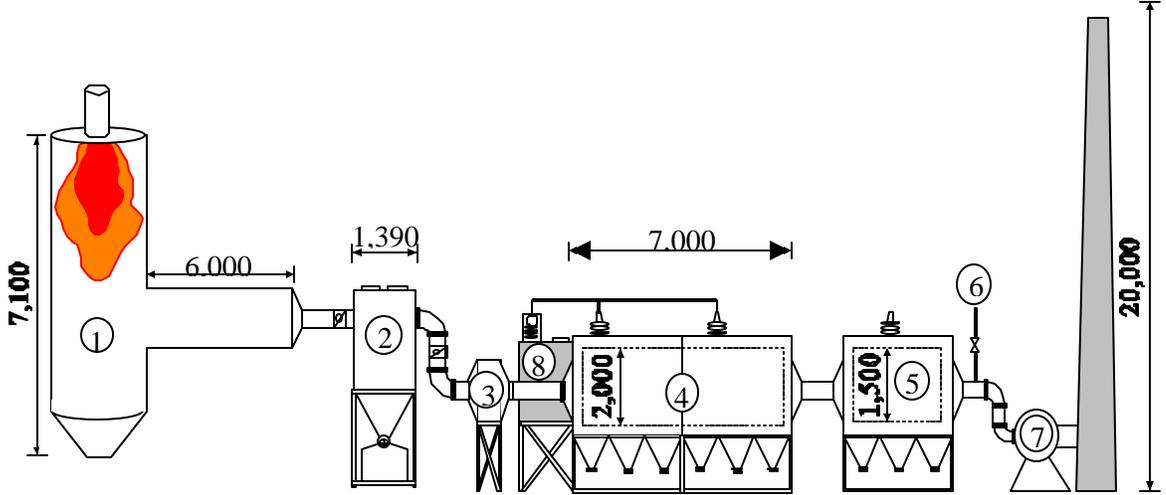


Fig 3. Layout of the industrial scale pilot plant installed at HANJUNG. ① Coal combustor, ② Bag filter, ③ Heat exchanger, ④ Pulsed corona reactor, ⑤ EP, ⑥ Sampling port for measurement, ⑦ ID fan, ⑧ Pulse generator.

III. Results and Discussions

1. Dummy load test

The matching between the pulse generator and the reactor is a crucial point to reduce electrical energy loss and to optimize the formation of streamer coronas [2]. Therefore, the output impedance of the generator should match that of the reactor in order to obtain the maximum transfer of energy to a load.

The generator was tested to find out the characteristics of its output impedance and a maximum transfer condition of output pulse energy to a load. Different non-inductive resistors (Carborundum Co.) were used as a load and pulse repetition rate was 1 pps.

With 30 kV of HVDC output, the maximum energy transfer occurs at 35 °C, as shown in Fig. 4. The transferred energy and efficiency to the resistor per pulse were 49 J and 68 %, respectively. Transfer efficiency has the same trend as Fig. 4. The efficiency of the generator and the energy per pulse were calculated using

$$\eta(\%) = \frac{E_L}{\frac{1}{2}C_0 \cdot V_{DC}^2} \times 100 \quad (2)$$

$$E_L = \int_0^t V(t)I(t)dt \quad (3)$$

where, C_0 : the capacitance of the initial capacitor
 V_{DC} : the output voltage of the HVDC supply
and τ : the pulse duration.

The power to a load is obtained by multiplying the energy per pulse dissipated in the

load by the pulse repetition rate. Fig. 5 shows the current and voltage waveforms versus time for 35Ω . The pulse voltage has 70.32 kV_{pk} , 280 ns rise time, and 640 ns duration. The current signal has 1.51 kA_{pk} , 220 ns , and 630 ns .

The impedance of discharge was computed with waveforms acquired by the oscilloscope as a function of time. All through the experiments, it is found that there is a constant impedance region and the value of impedance at the

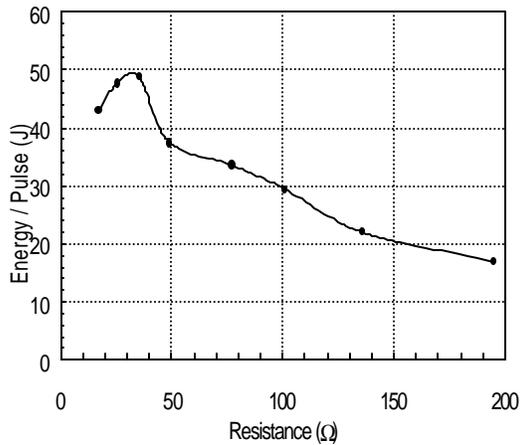


Fig 4. The transferred energy into the dummy loads.

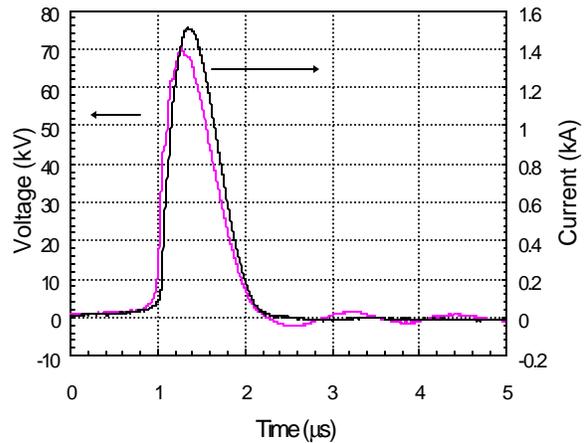


Fig 5. Voltage and current waveforms On the 35Ω dummy load.

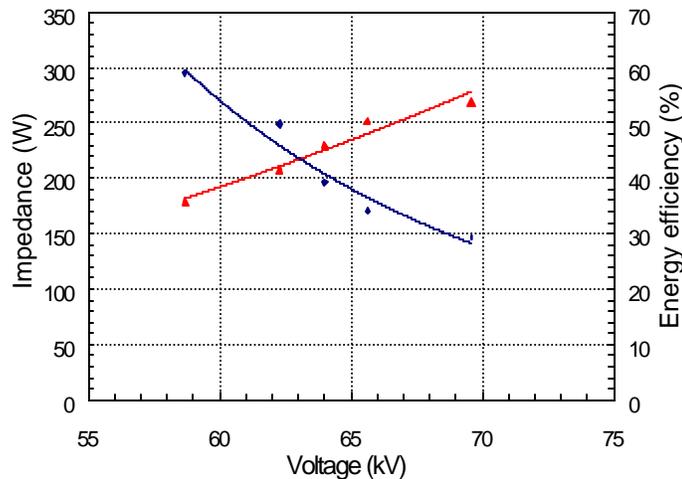


Fig 6. Impedance of the reactor (◆) and energy transfer efficiency (▲) as a function of the peak voltage applied.

region implies the minimum resistance of discharge. The minimum resistance is always roughly equal to the value of peak voltage divided by the peak current.

Fig. 6 shows a method to reduce impedance value of the reactor and to increase energy efficiency. With increasing the peak voltage applied, impedance of the reactor decreases and energy efficiency also increases.

2. Removal of SO₂

Fig. 7 shows the removal of SO₂ as a function of the molar ratio of NH₃ to SO₂ in flue gas. In this test, peak values of applied voltage and current and pulse repetition rate were 43.6 kV, 0.35 kA, and 90 pps, respectively. A variation of consumption of the electrical energy was constant during SO₂ removal tests.

Without injection of NH₃, the removal rate was only about 8 %. In general, the efficiency of SO₂ removal by a gas phase radical reaction is known only to be about 10 %. Although the removal process of SO₂ is mainly governed by the thermochemical reaction with ammonia and the heterogeneous chemical reaction, it can be enhanced by the pulsed streamer corona process [11]. When only NH₃ was injected into the gas with a 2:1 molar ratio to the initial SO₂ concentration, about 294 ppm of the initial 300 ppm SO₂ was removed. However, care must be taken with regards to secondary pollution production due to unreacted NH₃. During this experiment, emissions of NH₃ were detected up to 26 ppm. In addition, there must have been an unreacted additive even though we did not measure it. One of the methods that can reduce the emission of NH₃ as well as that of chemical additives is to modify the injection method or positions of NH₃ or chemical additives such as using a corona radical shower method [12].

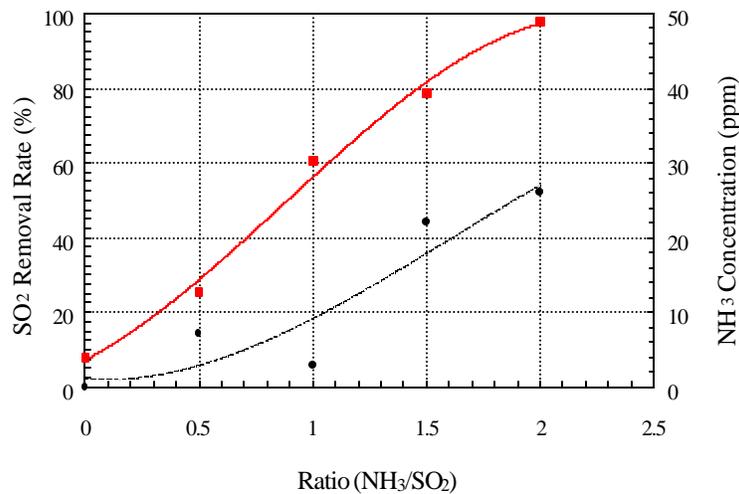


Fig 7. SO₂ removal rate(■) as a function of the molar ratio of NH₃ to SO₂ and emission of unreacted NH₃(●) at outlet of the reactor. The gas consisted of 18 % O₂ and 38 ppm CO. The flow rate and temperature of the gas were 1,200 Nm³/h and 118 °C, respectively.

3. Removal of NO_x

The tests were carried out to clarify the effect of the hydrocarbon additive, C₂H₄, and the energy input into the reactor on NO_x removal efficiency. Fig. 8 shows the removal rates of NO, NO₂, and NO_x with and without the addition of C₂H₄ in flue gas. The initial concentration of NO_x was (a) 245 ppm and (b) 312 ppm, and the voltage pulses

having a peak value of 85 kV were applied to the reactor.

As shown in Fig. 8 (a), in the case of no additive, the removal rate of NO increases with increasing the repetition rate of pulse but that of NO₂ decreases. This is because NO are oxidized to NO₂ by O and O₃ in plasma chemistry. Even though there is a difference between removal rates due to the higher or lower level of initial NO concentration, the removal characteristics of NO_x are similar when no additives are injected.

Fig. 8 (b) shows that the addition of C₂H₄ greatly enhances the removal efficiency of NO_x up to 52 % with nearly the same energy input as Fig. 8 (a). This results from OH and HO₂ radicals, which can be produced by C₂H₄ or hydrocarbon additives in the corona discharge process. NO and NO₂ are mainly converted into HNO₂ and HNO₃ by those radicals. The removal rates of NO, NO₂, and NO_x also increase as the pulse repetition rate increases.

The above results indicate that the chemical additive could be very effective in reducing the energy cost for the removal of NO_x using a nonthermal plasma technique. For the same energy input, higher NO_x removal can be expected with higher additive concentrations. However, higher unreacted hydrocarbons will be produced at higher additive concentrations. It is, therefore, desirable to achieve NO_x removal using the least amount of additive at an acceptable energy consumption.

Fig. 9 shows the removal of NO_x as a function of pulse repetition rates and peak voltage applied. The flow rate of the gas was 1,100 Nm³/h and the temperature at the inlet of the reactor was 120 °C. 600 ppm C₂H₄ was added to the flue gas.

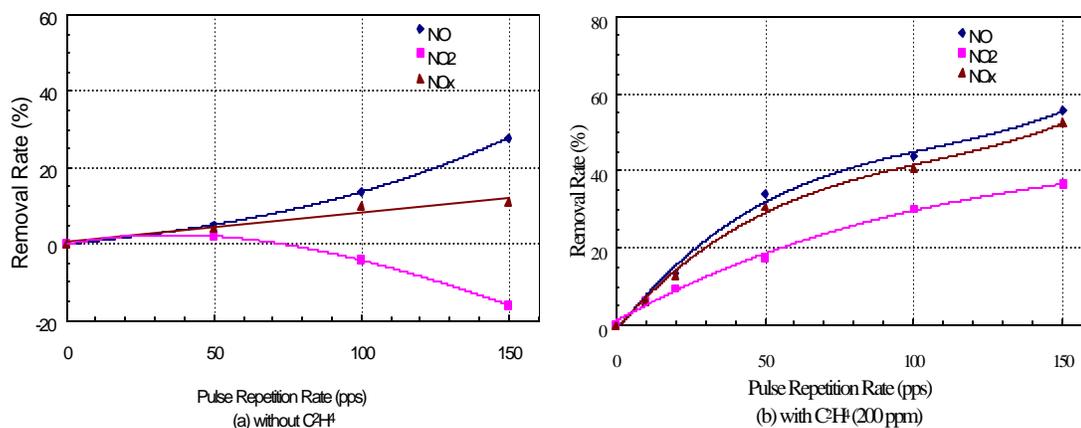


Fig 8. Effect of the additive and the input energy on NO_x removal. The gas consisted of 13 % O₂, 7 % CO₂, and 20 ppm CO. The flow rate and temperature of the gas were 1,200 Nm³/h and 120 °C, respectively.

By increasing the peak value of the pulse voltage from 60 kV to 90 kV, NO_x removal rates were 44.8 % to 63.6 % at 90 pps and 71.2 % to 78.6 % at 200 pps. The removal rate of NO_x increases with increasing the peak value of pulse voltage at lower pulse repetition rates but it has a saturation region. When a pulse repetition rate increases, the removal rates also increase but reaches a saturation point. Therefore, it is indispensable for consideration of economic aspects since the removal efficiency tends to saturate

owing to existence of the reverse reaction.

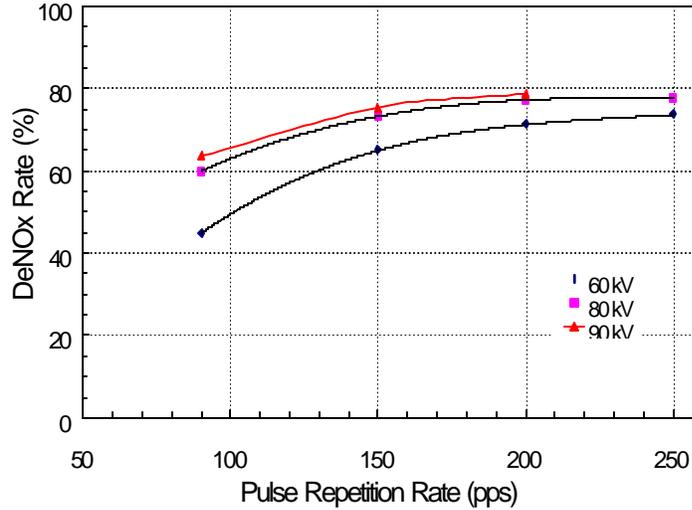


Fig 8. Effect of peak values and the input energy on NO_x removal rate. The initial NO, NO₂, and NO_x concentration were 218, 70, and 288 ppm, respectively.

About 79 % of the initial NO_x concentration was removed with an energy consumption of 9.23 Wh/Nm³. At that time, the pulse repetition rate was 200 pps and the delivered energy to the reactor was 51 J, 90.32 kV of peak voltage and 1.55 kA of peak current were measured. With reference to ENEL work [4], the NO_x removal was 50 % with initial NO_x concentration of 240 ppm and about 12 Wh/Nm³ of electrical energy was supplied to the gas.

IV. Conclusions

We have developed and tested an industrial-scale nonthermal plasma DeSO₂ and DeNO_x pilot plant composed of a wire-to-plate type reactor and a MPC type pulse generator.

As a part of energy saving, this paper reported the impedance characteristic and matching conditions between the pulse generator and the reactor, and showed some test results for the removal of SO₂ and NO_x with and without a hydrocarbon additive.

The results of the dummy load tests indicated that the output impedance of the pulse generator was about 35 Ω. Matching between a reactor and a pulse generator, the impedance of the reactor should be equal to that of the pulse generator when voltage pulses are applied to the reactor, if possible.

The experimental results obtained at 118 °C show that removal efficiency of SO₂ with a molar ratio of 2:1 is 98 % when compared to that of SO₂ of 8 % without NH₃. The addition of C₂H₄ significantly enhanced the performance of NO_x removal with up to 79 % with an energy consumption of 9.23 Wh/Nm³. But the removal efficiency of NO_x does not increase monotonically.

In conclusion, the present simultaneous DeSO_x-DeNO_x system using a nonthermal plasma method is found to be competitive with existing technologies.

Further research is needed, especially focusing on optimization to improve removal efficiency, to modify injection methods of an additive, and to reduce energy requirements. For example, the effect of gas residence time and geometry of a reactor on removal of NO_x and SO₂ should be investigated. Furthermore, the efficiency of a pulse generator should be higher and the size of the pulse generator should also be smaller.

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