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COMBINED TREATMENT OF SO_2 AND HIGH RESISTIVITY FLY ASH USING A PULSE ENERGIZED ELECTRON REACTOR

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A combined treatment which yeilds an acceptable removal of SO_2, NO_x, and particulate emissions from coal-fired boilers in a single compact system promises several advantages with regard to capital investment and operational cost. Synergistic effects between the particles and the gases in the removal process can further advance the economic feasibility. A system which can be retrofitted in existing electrostatic precipitator or baghouses would be especially attractive.

The combined removal of SO_2 and high resistivity fly ash has been demonstrated in a pulse energized electron reactor (PEER). The PEER system which was originally developed for the removal of SO_2 utilizes a positive pulse streamer corona discharge in a non-uniform field geometry.

Using a wire-to-cylinder geometry (cylinder: 10 cm inner diameter), and a pulse high voltage (+45 kV peak voltage, 200 ns width, 60 Hz repetition frequency), the PEER system removed more than 90 % of the SO_2 with a substantially smaller power requirement than that necessary for high energy electron beam treatment.

It is known that a high energy electron beam (400 - 800 keV) promotes chemical reactions which remove SO_2 and NO_x from stack gas. High energy electrons ionize and/or excite gas molecules to produce radicals (O, OH, OH_2, etc.). The radicals react with SO_2 and form aerosols which can be collected by an electrostatic precipitator or a bag filter. Since the formation energy of the radicals is in the range of 5 - 15 ev, they can be formed in a streamer corona discharge. The streamers produce electrons which promote chemical reactions by ionizing molecules, energizing excited molecular states, breaking molecular bonds, and forming free radicals.

When the PEER is operated with a pulse superimposed on a dc bias voltage (combined treatment operation of the PEER), it can collect high resistivity dusts more efficiently than a conventional ESP with a dc-only operation. A back corona free condition can be obtained during the pulse off period even if the collecting electrode is covered with a high resistivity layer. The positive streamers formed in the pulse on period travel across the electrode spacing to the collecting electrode, and trigger breakdowns of the dust layer and neutralize the surface charge. Positive ions are left by the streamers, and are driven by the dc field during the pulse off period. The quantity of these ions accumulating on the surface of the neutralized dust layer can be controlled to be below a threshold level of a back corona initiation by adjusting both the pulse voltage and the dc bias voltage. The positive ions form an ionic current and charge the suspended dust particles. The charged particles are then driven to the collecting electrode during the pulse off period.

The fly ash particle collection efficiency (η_p) of the PEER in the combined treatment operation was measured. The η_p for a dc-only operation (simulation of a conventional ESP) was also measured. When the collecting electrode was covered with a high resistivity layer, the η_p of the PEER in the combined treatment operation was significantly better because an intense back corona took place in the dc only operation. Measurement of fractional collection efficiency indicated that the PEER agglomerated the fly ash. In the combined treatment operation monopolarly charged particles were observed, while in the dc-only operation small particles were charged with both polarities and many neutral particles were observed. It is noted that the SO_2 removal efficiency is improved by the addition of fly ash.

These results indicate that the PEER can be used as the combined removal of SO_2 and fly ash. The electrode configulation and performance results make retrofit consideration attractive.

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ABSTRACT

The combined removal of SO_2 and high resistivity fly ash has been demonstrated in a pulse energized electron reactor (PEER). The PEER system which was originally developed for the removal of SO_2 utilizes a positive pulse streamer corona discharge in a non-uniform field geometry. In performance tests on SO_2, more than 90 % was removed with an advantageously small power requirement. Combined treatment performance was demonstrated by introducing high resistivity fly ash into the test gas and the PEER is significantly more efficient than a conventional electrostatic precipitator operated with a dc voltage. Observations show that the PEER agglomerates the fly ash and further that the SO_2 removal efficiency is improved by the presence of fly ash. The electrode configuration and performance results make retrofit consideration attractive.

1. INTRODUCTION

A combined treatment which yeilds an acceptable removal of SO_2, NO_x, and particulate emissions from coal-fired boilers in a single compact system promises several advantages with regard to capital investment and operational cost. Synergistic effects between the particles and the gases in the removal process can further advance the economic feasibility. A system which can be retrofitted in existing ESP or baghouses would be especially attractive.

A pulse energized electron reactor (PEER) has been developed for the removal of SO_2 from gas streames.(1) The PEER uses a pulsed streamer corona discharge in a non-uniform electrode geometry. Using a rod-to-plane or a wire-to-cylinder geometry, the PEER removed more than 90 % of the SO_2 with a substantially smaller power requirement than the dc discharge or the electron beam processes. Since the PEER can also be used with a wire-to-plate geometry, retrofitting a conventional ESP for SO_2 removal using pulsed streamer corona may be simplified. The possibility of using the PEER for the combined removal of SO_2, NO_x, and particles depends on the particle removal performance of the PEER.

In the current study, the particle collection efficiency (η_p) of the wire-to-cylinder geometry PEER was measured using fly ash. The PEER was operated with dc power only to simulate a conventional ESP and with pulsed power and dc bias in the combined treatment operation of the PEER. The collection efficiency was measured using fly ash particles and using a collecting electrode covered with a high resistivity layer of filter paper to simulate back corona in fly ash cake. The fractional particle collection efficiency of the PEER was also determined using an optical particle counter. The charge-to-radius ratio distribution of the particles exiting the PEER was measured. The effect of fly ash on the SO_2 removal efficiency of the PEER was investigated.

2. OPERATIONAL CONCEPT OF THE PEER

2.1 SO_2 removal

It is known that a high energy electron beam (400 - 800 keV) promotes chemical reactions which remove SO_2 and NO_x from stack gas.(2) High energy electrons ionize and/or excite gas molecules to produce radicals (O, OH, OH_2, etc.).(3) The radicals react with SO_2 and form aerosols which can be collected by an electrostatic precipitator or a bag filter.

Since the formation energy of the radicals is in the range of 5 - 15 eV, they can be formed in a streamer corona discharge. The streamers produce electrons which promote chemical reactions by ionizing molecules, energizing excited molecular states, breaking molecular bonds, and forming free radicals. The PEER has a non-uniform electtrode geometry and utilizes short pulses of high voltages.

The PEER reactor chamber used in this experiment is depicted in Fig. 1, and the voltage wave form is shown in Fig. 2. Using short duration voltage pulses allows a very high electric field to be established without sparkover, and results in the formation of more intense and uniform streamers. Using positive polarity instead of negative causes the streamers to reach out farther to cover a large volume. The pulsed operation of the PEER is very power efficient since almost all the current is formed by electron migration and power loss during the pulse and due to driving ions is small.

Using a pulse voltage superimposed on dc-bias results in a high peak voltage which increases the streamer intensity. The appropriate dc-bias level is determined by optimizing the overall performance taking into account the power wasted by dc driven ions.

2.2 High resistivity fly ash removal

The PEER is operated with a pulse superimposed on a dc-bias voltage to collect suspended particles in flue gas. Positive polarity is used to remove SO_2 at the same time, and the dc voltage is set at a value less than the dc corona starting voltage to prevent back corona when the dust resistivity is very high. There are two modes of operation.

(1) Pulse on --- ash cake discharge
The following process for ash cake discharge is proposed as an explanation for the good performance with a layer of high resistivity fly ash on the plate electrode. During the pulse (see Fig. 2(A)), positive streamers travel across the electrode spacing to the collecting electrode which is covered with a high resistivity of filter paper which replicates fly ash cake under back corona conditions. Since the streamer channel is conductive and can be regarded as an extention of the discharge electrode, the streamers approaching the simulated dust layer produce a high electric field locally between the tip of the streamers and the insulating layer. The electric field at the tip of the streamers where electron avalanches are taking place is also very intense. The high electric fields due to the approaching of streamers trigger breakdowns of the dust layer. The charges accumulated on the high resistivity dust layer are released by these breakdowns and the surface of the layer is neutralized. Electrons and positive ions are produced in the streamers. Electrons (high mobility) move to the discharge electrode through a conductive streamer channel, and the positive ions (low mobility) are left along the channel after the pulse voltage is removed.

(2) Pulse off --- particle charging and collection
The positive ions left by the streamers are driven by the dc field. These form the ionic current

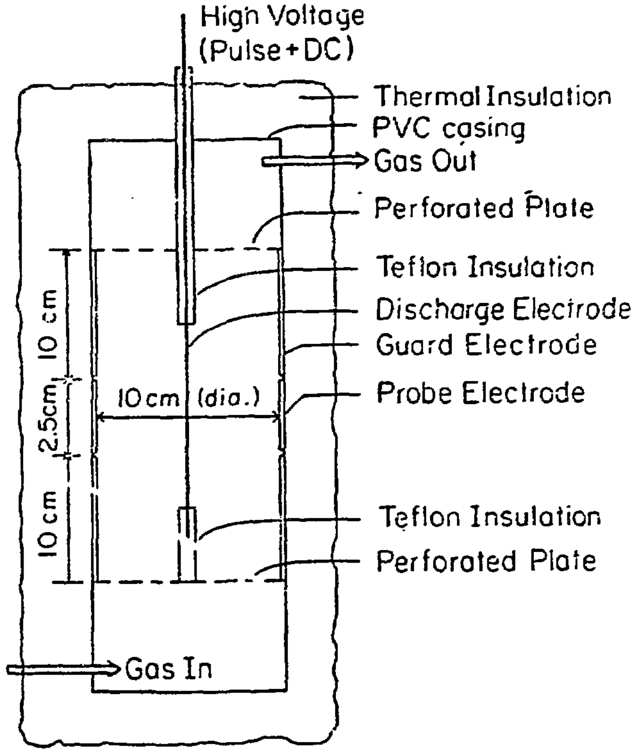
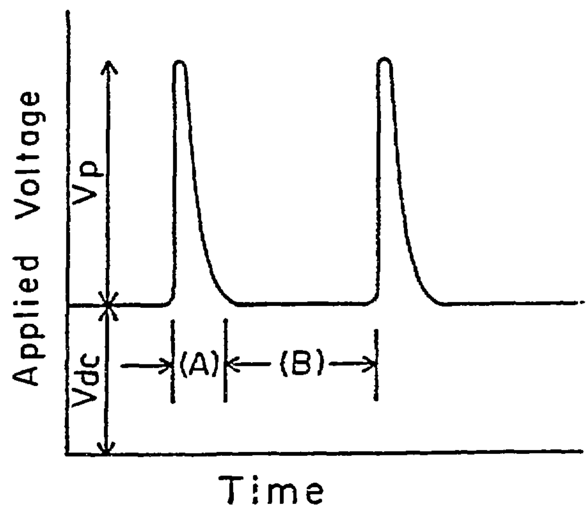


Fig. 1 The PEER chamber



(A) Pulse on period
(B) Pulse off period

Pulse width: 200 ns
Pulse frequency: 60 Hz
Pulse voltage: +40 ~ +45 kV
DC voltage: adjustable

Fig. 2 Applied voltage waveform

and charge the suspended particles during the pulse off period (see Fig. 2(B)). The charged particles are then driven to the collecting electrode by the dc field. When the dust resistivity is very high, the charge carried by the ions and the charged particles accumulates on the surface of the dust layer. This surface charge is then neutralized during the next pulse on period. The surface charge density can be controlled to be below a threshold value for the back corona initiation(4) by adjusting the pulse voltage, frequency, and the dc-bias voltage. This control provides the back corona free condition during the charging and collection (pulse off) period.

3. EXPERIMENTAL APPARATUS

3.1 The PEER chamber and flow diagram

The PEER chamber used in these experiments is shown in Fig. 1. The wire-to-cylinder electrode is made of stainless steel to prevent corrosion. The cylindrical collecting electrode (inner diameter = 10 cm) has a current measuring electrode (length = 2.5 cm) at its center, and the wire electrode (diameter = 3 mm) was covered with braided cable to increase the roughness of its surface. The length of the wire electrode was either 5 cm or 10 cm, and both ends were covered with teflon insulation to prevent the streamer corona. The electrode assembly was encased in PVC (polyvinyl chloride) tubing.

The flow diagram is shown in Fig.3. In the SO_2 removal experiment, air containing SO_2 (1000 ppm) and water vapor 2.5 % by volume (100 % RH) was used at temperatures of 22 °C and 110 ± 10 °C. In the fly ash collection experiment, dry air (less than 8 % RH and without SO_2) was used at 22 °C. Fly ash with a mean diameter of 5 ~ 10 μm was added using a fluidized bed. The resistivity of the fly ash was 1.0 × 10^{13} ohm-cm under these conditions. The gas exiting the PEER was diluted with dry air and then analyzed. The SO_2 concentration was measured by a pulsed flourescent SO_2 analyzer and the fly ash concentration was monitored by an optical particle counter.

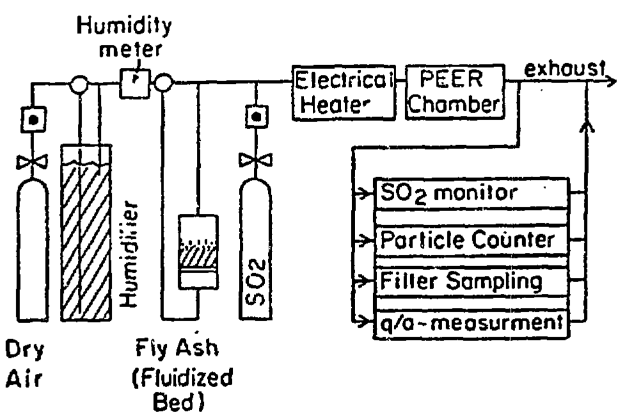


Fig.3 Experimental flow system

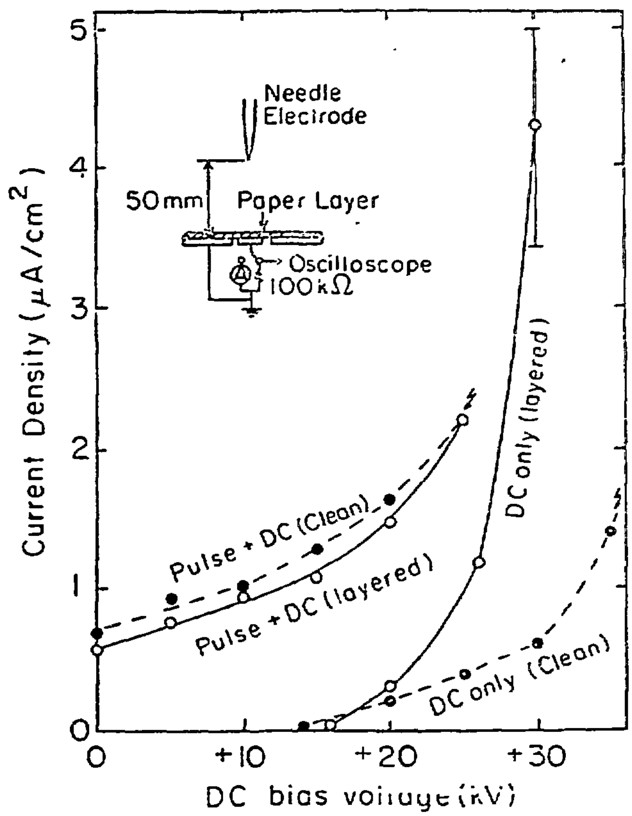


Fig.4 Voltage-current characteristics (needle-to-plate electrode)

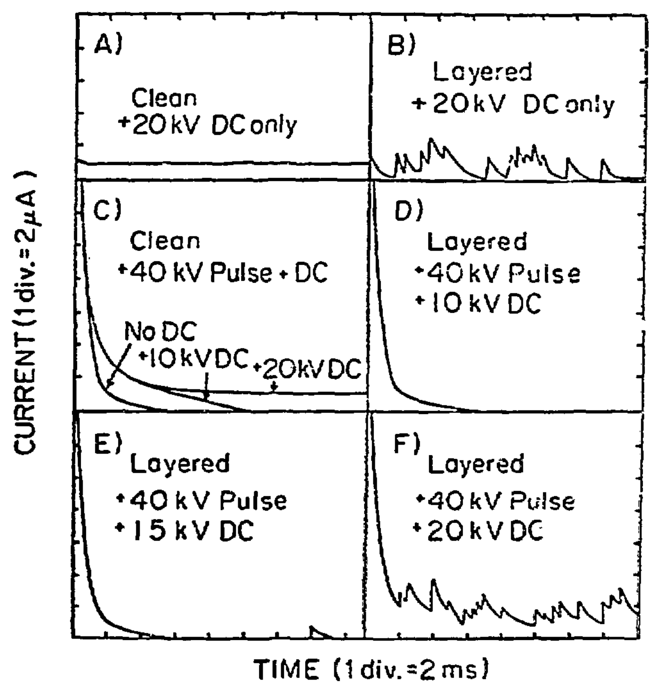


Fig.5 Current waveform

3.2 Current waveform measurement

In order to verify that the PEER can operate free of back corona during the pulse off period, the current waveform was measured using a needle-to-plate electrode as shown in Fig. 4. The electrode separation was 50 mm, and radius of curvature at the tip of the needle electrode is approximately 0.1 mm. A 1.0 mm thick layer of porous tissue paper was placed on the plate electrode to trigger back corona. The electrode is placed inside a chamber containing dry air to increase the resistivity of the paper to 1.0 × 10^{14} ohm-cm. A storage oscilloscope and a 100 k-ohm resistor were used as shown in Fig. 4 to measure low level ionic current. The rise time of the current waveform measurement circuit was about 200 μs.

3.3 Pulse voltage source

Pulse voltage is generated using a capacitor bank and a rotating spark gap.(5) A dc bias voltage can be superimposed on the pulse using a coupling capacitor. The output pulse voltage used in this experiment had a peak value range of +40 to +45 kV, a 200 ns width, and a 60 Hz repetition frequency.

4. EXPERIMENTAL RESULTS

4.1 Current waveform measurement

Using the needle-to-plate electrode, V-I curves and current waveforms were measured with and without a high resistivity paper layer on the plate electrode. Figure 4 shows the voltage and current characteristics. Slightly above the dc corona starting voltage of +14 kV, a large current increase due to back corona was observed using dc voltage only and a 10^{14} ohm-cm paper layer (1 mm thick). When the pulse (+40 kV peak voltage) and dc bias voltage < +14 kV was used with the paper layer, the current was slightly smaller than that with a clean electrode. This indicates that back corona was not occurring and that the high resistivity layer reduced the intensity of the streamer corona. As the dc bias voltage was raised above the dc corona starting voltage, the current with the paper layer increased rapidly. It became the same as that of the clean electrode at a dc level slightly below sparkover, suggesting that a weak back corona took place.

Figure 5 (A, B) shows the current waveforms using dc only (+20 kV). Without the high resistivity layer, a smooth dc current with only a small fluctuation was observed. With the layer (at the same dc voltage of +20 kV) the current waveform contained pulses which are due to back corona.(4)

Figure 5 (C,D,E,F) shows the current waveform using the pulsed power and dc bias voltage. The pulses in this measurement had a +40 kV peak voltage, a 200 ns pulse width, and a 60 Hz repetition frequency. Figure 5 (C) is the current waveform using a clean electrode without the high resistivity layer. At a dc bias of +10 kV, an increase in the current at the tail of the pulse current waveform was observed. This is probably an increase in the current carried by ions which were produced by the previous streamers. When the dc bias voltage was +20 kV and exceeded the dc corona starting voltage (+14 kV), a continuous current was observed between the pulses for the clean electrode condition. Figure 5 (D,E,F) is the current waveform with the high resistivity layer. At dc bias voltages below +14 kV, no back corona pulses were detected (see Fig. 5 (D)), but at +15 kV dc bias voltage, small current pulses due to back corona were observed at the tail of the current waveform (Fig. 5 (E)). Above +15 kV dc bias voltage (Fig.5 (F)), many large current pulses due to back corona were observed. Since the layer resistivity was very high (1.0 × 10^{14} ohm-cm), back corona was immediately triggered by the presence of the dc corona (which occurred when the dc bias voltage was higher than the dc corona starting voltage).

These results show that, during the pulse off period, a back corona free condition can be obtained in the PEER by adjusting the dc bias voltage even when the electrode is covered with a very high resistivity layer.

4.2 SO_2 removal performance

The SO_2 removal efficiency of the PEER is shown in Fig. 6. A pulse of +45 kV peak voltage, 200 ns width, and 60 Hz repetition frequency was used with a dc bias voltage. The test gas containing 1000 ppm SO_2 and 2.5 vol% H_2O was used at a temperature of 22 °C with a flow rate of 4.1 ℓ/min. The exposed length of the discharge electrode of the PEER was 5 cm. Since the starting point of the streamer corona is limited to

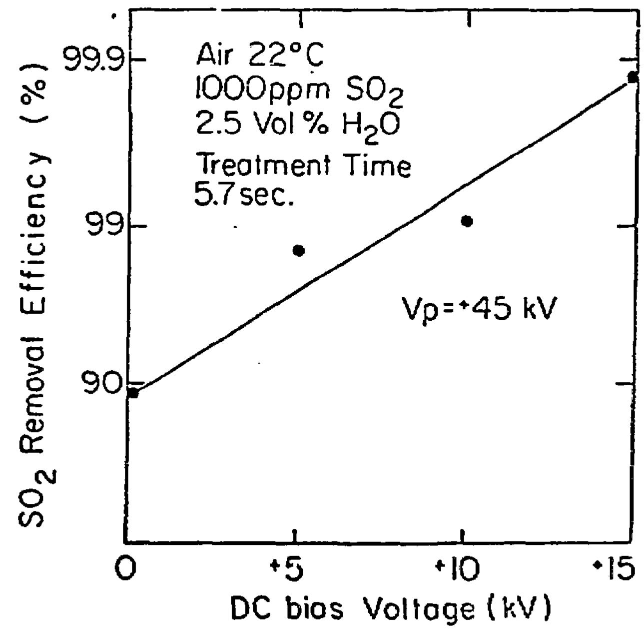


Fig. 6 SO_2 removal efficiency

the exposed metal surface of the discharge electrode and the streamers do not significantly expand axially beyond the discharge origin, the length of the gas treatment volume is considered to be the same as that of the discharge electrode. Therefore the gas treatment time is assumed to be 5.7 sec at a gas flow rate of 4.1 ℓ/min. The SO_2 removal efficiency becomes higher at larger dc bias voltages. A white powder was observed covering the discharge electrode after operation of the PEER. The white powder and a mist condensation were also observed on the collecting electrode.

The SO_2 removal efficiency of the PEER at higher temperatures is tabulated in Table 1. A pulse of a peak voltage V_p = +45 kV was used with a dc bias of V_{dc} = +10 kV. The gas treatment time was 5.7 sec at 22 °C, 4.8 sec at 80 °C, and 4.4 sec at 110 °C. The SO_2 removal efficiency decreases at higher temperatures. This is due to the shorter treatment time as well as to an increase in the rate of the back reaction in the SO_2 removal process. Such an increase in the back reaction was reported in the electron beam process for SO_2 removal.(3)

In order to increase the SO_2 removal efficiency and promote the formation of solid products, NH_3 can be added to the flue gas. The power requirement of the PEER was smaller than that for the high energy electron beam process as reported separately.(1)

TABLE 1 SO_2 removal efficiency of the PEER at high gas temperatures

Temperature (°C)	SO_2 removal efficiency (%)	Gas treatment time (sec)
22	99	5.7
80	94	4.8
110	80	4.4

4.3 SO_2 removal (with fly ash)

Previous electron beam treatment work showed that the presence of white carbon powders in flue gas improves the NO_x removal efficiency.(6) Unstable radiochemical products from NO_x condense on the surface of the powders, and are stabilized by a reaction with the absorbed water on the powders. This reduces the rate of the back reaction and improves the removal efficiency. Similar reduction process of the back reaction is expected for the SO_2 removal by the PEER.

The SO_2 removal efficiency of the PEER was measured with and without suspended fly ash particles. A fly ash with a mean diameter of 5 - 10 μm was used at a dust concentration of 4 g/m³. The experiment was conducted using the test gas with 1000 ppm SO2 and 2.5 vol% H_2O at room temperature (22°C). The gas treatment time was reduced to 3.2 sec for the comparison of the SO_2 removal efficiency with and without fly ash. A +45 kV pulse peak voltage with a +15 kV dc bias was used. The 55 % SO_2 removal efficiency (45 % penetration) was improved to 86 % (14 % penetration) by the presence of the fly ash particles.

4.4 Fly ash removal performance

In order to simulate a high resistivity dust layer, a 1 mm thick paper layer was attached to the collecting electrode of the PEER. The length of the discharge electrode was 10 cm and the gas treatment time was 3.9 sec. Dry air at 22 °C was used at a flow rate of 12.0 ℓ/min with a fly ash concentration of 4 g/m³. The fly ash concentration was measured at the exhaust of the PEER. The fly ash removal efficiency was determined from the ratio of the fly ash concentrations with and without the voltage application, so that mechanical collection was not included in the determination of removal efficiency.

The voltage-current characteristics are shown in Fig. 7. When the PEER was powered by a dc bias voltage only (simulation of a conventional ESP with dc operation), a large current increase and the typical hysteresis due to intense back corona were observed. When the dc voltage was raised, the dc corona started at +20 kV and back corona took place immediately.

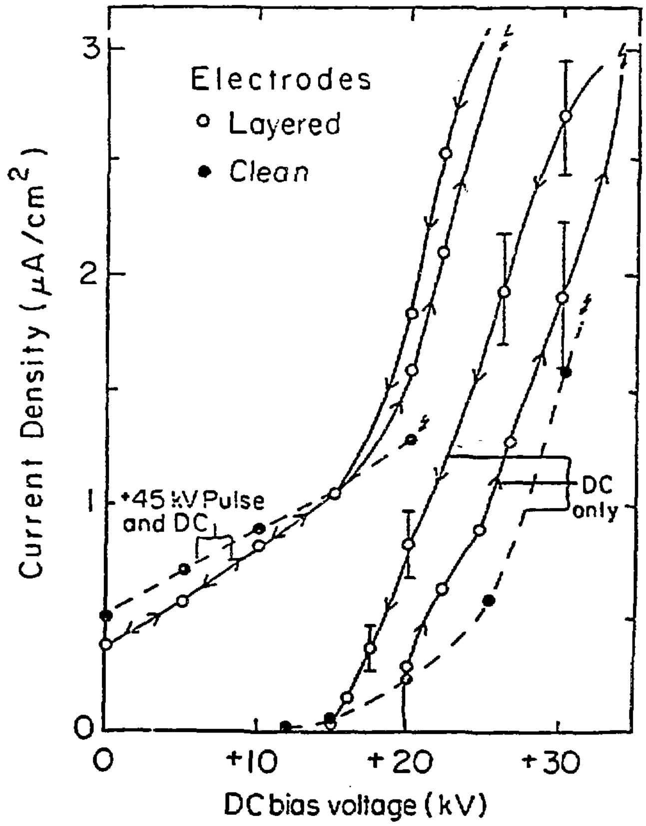


Fig. 7 V - I characteristics of the PEER (wire-to-cylinder electrode)

The current increased very rapidly. Then the dc voltage was decreased, and the current was quenched at +15 kV. Figure 7 also shows the V-I characteristics using the pulse (V_p = +45 kV) and a dc bias voltage (combined treatment operation of the PEER). Below V_{dc} = +15 kV, a large current increase was not observed, suggesting that no back corona took place. Above +15 kV, the current increased rapidly and the hysteresis due to back corona was observed.

The overall collection efficiency of the PEER was measured using pulse voltage with a +15 kV dc bias voltage, so that a back corona free condition was achieved in the period between the pulses. The collection efficiency (shown in Table 2) was obtained from the difference in weight gained by a filter sampling fly ash at the exhaust with the PEER on and off. The collection efficiency using only dc power (+25 kV) was also measured. A 50 % collection efficiency (50 % penetration) was achieved using the PEER powered by the pulse and dc bias voltage, while for the dc only operation the collection efficiency was 7 % (93 % penetration). The lower collection efficiency in the dc only operation (with severe back corona) is plausible since mechanical collection was not included in the efficiency calculation.

TABLE 2 Fly ash collection efficiency

Condition	Penetration (fly ash concentration) (g/m^3)	Collection efficiency (%)
No voltage	1.88	0
PULSE + DC V_p = +45 kV V_{dc} = +15 kV J = 1.0 $\mu A/m^2$	0.94	50
DC only V_{dc} = +25 kV J = 1.0 $\mu A/m^2$	1.75	7

V_p: pulse peak voltage
V_{dc}: dc bias voltage
J: current density

The fractional penetration of fly ash particles was also measured under the same condition using an optical particle counter (see Table 3). The penetration of the smaller particles (< 5 μm diameter) decreased to less than 10 %, while that of the larger diameter particles increased. The increase in the penetration of the larger particles suggests an agglomeration of particles due to reentrainment. The penetration of particles in the 3 - 5 μm range for the pulse with dc bias operation was about 1/2 that for the dc only operation. In the pulse with dc bias operation, reentrainment can result from streamers hitting the dust layer during each pulse period. In the dc only operation,

TABLE 3 Fractional penetration of fly ash

Fly ash diameter range (%)	Penetration (%)	
	DC only V_{dc} = +25 kV	PULSE and DC V_p = +45 kV V_{dc} = +15 kV
2 - 3	7	7
3 - 5	10	6
5 - 10	25	17
> 10	100	400

reentrainment occurs because of back corona.

The charge-to-radius ratio (q/a) of the fly ash particles was also measured.(7) A sampling tube was inserted into the PEER chamber with the sampling point 2 mm above the measuring electrode. The q/a distribution of a sample of 50 particles (approximately 1 μm diameter) is shown in Fig. 8. In the pulse with dc bias operation the 1 μm diameter particles were charged monopolarily, while in the dc only operation the particles were charged with both polarities and many neutral particles were observed. The q/a of the larger particles was small in both operations, suggesting that reentrainment had occurred.

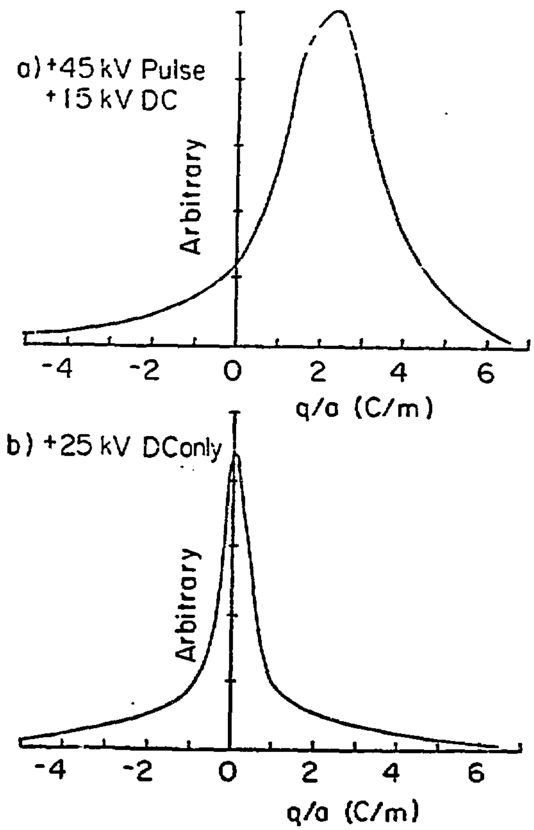


Fig. 8 Charge-to-radius ratio distribution of fly ash particles (Diameter: approx. 1 μm)

5. CONCLUSION

A pulse energized electron reactor (PEER) has been developed which removed SO_2 and high resistivity dust from gas streams in a combined treatment by one device. The performance of the PEER was investigated using a wire-to-cylinder electrode and the following conclusions were obtained.

(1) The PEER can remove more than 90 % of the SO_2 from a gas stream, and the removal efficiency is improved by the presence of fly ash particles in the gas stream.

(2) The collection efficiency of fly ash particles by the PEER is significantly higher than that by a dc powered precipitator when the collecting electrode is covered with a high resistivity layer.

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