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THE USE OF ENERGETIC ELECTRONS IN A PARTICLE PRECHARGER AND IN A SULFUR DIOXIDE REACTOR*

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ABSTRACT

Electrons energized by corona discharge to produce the necessary ionization for particle charging have been used in electrostatic precipitators for decades. This paper reports the use of an electron beam to release and energize electrons which produce copious charging currents in a bench precharger. Results have been obtained for various values of electron beam energy, beam current, electric field strength, current density, and exposure time in measurements of charging efficiency for large conducting spheres and 1 and 3 μm diameter PSL particles. After matching the beam energy and geometry in the bench precharger, particle charges greater than five times the theoretical ionic charging value were measured in the bi-electrode precharger. The increased charge can be explained by space-charge enhancement of the electric field and/or free electron charging. The use of very high energy electron beams for the removal of SO_2 and NO_x from flue gases has been previously established elsewhere. Since the energy regime for the electrons required for the production of oxidizing radicals is the order of 10 eV, a device which operates in a lower energy regime is attractive. A positive streamer corona device has been constructed and used to energize electrons for the purpose of producing oxidizing radicals. The performance of a pulse energized electron reactor (PEER) has been evaluated. More than 90% of the SO_2 has been removed from a test gas stream containing air, water vapor and 1,666 ppm of SO_2. The power efficiency of the PEER device is greater than that for DC discharge or high energy electron beam treatment.

INTRODUCTION

Energetic electrons have been used in the technology of flue gas clean-up for decades, but until recently the energization process has been primarily that of corona discharge in electrostatic precipitators (White, 1963). The energies of the electrons are those produced by a corona discharge and the purpose of the energetic electrons is to supply the necessary ionization to provide ion or electron currents which in turn charge particles for removal by means of electric fields. In a very different energy regime, energies of several hundred thousand electron volts up to one million electron volts, Japanese scientists showed that chemical reactions promoted by the energetic electron flux effectively converted SO_2 and NO_x (Tokunaga, 1978; Kawamura, 1981) into readily removable products. Two commercial projects using energetic (400-800 keV) electrons are underway in the United States at this time (Williams, 1983).

The choice of the energy regime of electrons to be used in flue gas clean-up processes is an important factor in performance efficiency and in power consumption efficiency. The optimization of electron energy will in turn dictate the choice of energizing mechanism. The possibilities now include:

1. Electron beams of energies matched to the desired application
2. DC discharges
3. Pulsed discharges of both polarities
4. [illegible]ave energization
5. [illegible] speed gas discharges
6. [illegible]oto absorption methods.

The two examples of energetic electron applications presented in this paper are an energy-geometry matched particle precharger and a positive pulse energized electron reactor (PEER).

The particle precharger takes advantage of the copious ionization produced by energetic electrons (Finney, 1981; Clements, 1981). The

electron beam precharger test facility. The Florida State University has been previously described (Clements, 1983). The early tests with this system showed that the electron beam energy must be selected to appropriately match the electrode geometry of the precharger. The first reports have been presented elsewhere (Clements, 1984; Mizuno, October, 1984).

The development of the pulse energized electron reactor (PEER) which utilizes positive pulse streamer corona to energize electrons resulted from a search for a power efficient means of radiation dose enhancement (Davis, 1982). Experiments with secondary ionization showed that dose enhancement factors of 3 or 4 could be readily achieved by DC electric fields but that an intolerably large power consumption was required. By positive pulsed streamer corona energization, electrons are efficiently energized while at the same time, dissipative ionic currents are minimized.

A long term objective of the project is the combined removal of SO_2, NO_x and particulate matter in an integrated system. The results for the removal of SO_2 and particulate matter in a modified PEER device are presented in a separate paper at this conference (Mizuno, November, 1984).

PARTICLE PRECHARGER USING ENERGETIC ELECTRONS

The electron beam precharger test facility has been previously described (Finney, 1983; Clements, 1983). The system shown in Fig. 1 is designed for the flexible tests of precharger modules using electron beam ionization. In the first test, an approximately 1 MeV electron beam was used in the Mark I precharger shown in Fig. 2. The importance of matching the beam energy with the electrode geometry was immediately evident in that spurious ionization took place outside the charging zone and indeed outside the confines of the precharger itself. However copious the ion currents, the final charge on the particle is determined by the ratio of the positive and negative ion populations. The lowest energy available from the Van de Graaff accelerator was too large for the geometry of the particle precharger. This is not an intrinsic disadvantage of electron beam utilization but rather means that large prechargers, commensurate with field application, are appropriate for electron energies of the order of 1 MeV.

To achieve proper energy geometry matching as schematically shown in Fig. 3 and to utilize the test system which was already as large as could be accomodated in the laboratory, a low energy electron source was needed. The source constructed for this purpose is shown in Fig. 4. The source produces electrons in the energy range 0-100 keV in the current range of 0-10 μA.

To flexibly confirm energy geometry matching and measure particle charges produced by electron beam ionization, a tri-electrode system was constructed outside the confines of the test system as shown in Fig. 5. The results with the tri-electrode system are shown in Fig. 6. The test particles were 1.1 μm diameter PSL particles. The solid points in Fig. 6 are for air while the open point is for nitrogen. Dashed lines give the values for saturation charge computed with Pauthenier's theoretical expression with two values for the dielectric constant. A dielectric constant of infinity corresponds to a conducting particle while the lower line is for a value of 2.5, that for PSL. The conclusion is that the Pauthenier limit is at least achieved in the tri-electrode system operating on air.

The point for the suspension in nitrogen in Fig. 6 lies above the data and calculations for air. Presumably the improved charge is due to free electron charging which can take place in nitrogen because of the absence of electronegative gases. Only one value is shown for nitrogen since for higher charging electric fields there is evidence that the particles were sufficiently charged to self-precipitate and not reach the charge measurement system, a q/a device which was developed by Mizuno (Mizuno, 1982).

A series of measurements were made with a bi-electrode precharger which is schematically shown in Fig. 7. Here again, the ionization zone is strictly limited by the range of the electrons in air, approximately 10 cm. Monopolar charging takes place in the charging zone under the action of the electric field due to the potential difference between the grid electrode and the plate electrode. The results of charging measurements using 1.1 μm diameter PSL particles is shown in Fig. 8. The measured particle charge is substantially above the range of calculated values using Pauthenier's theory and approximately five times larger than that for the charge calculated with the dielectric constant for PSL. A part of the explanation of the large measured charge is the fact that the ionization zone reduces the spacing between the electodes and increases the electric field for a given potential difference. This accounts for only a part of the improvement. The remainder is presumably due to an increase in the free electron population due to the increased electric field.

The charge deposited on 3.0 μm diameter particles was measured in the bi-electrode system as shown in Fig. 9. An increase in particle charge is observed but the increase is not as marked as that for 1.1 μm diameter particles. Additional experiments are needed to establish the systematics of particle size and determine the relative roles of the several charging mechanisms.

R.H. Davis

In preparation for these experiment studies of the control of the ionization zone and the charging zone were carried out with drop shot equipment and conducting spheres suspended by insulating filaments (Clements, August, 1984). The studies confirmed on a macroscopic scale the need to match the electron energy (range) with the design spacial extent of the ionization zone. The results are incorporated in the Mark II precharger (Fig. 10) which is now under test.

A PULSE ENERGIZED ELECTRON REACTOR FOR SULFUR DIOXIDE REMOVAL

Scrubbers are the most commonly used devices for the removal of sulfur dioxide from stack gas emitted by coal-burning power plants. While scrubbers are effective, their capital cost and operating expense motivated a search for alternate means of SO_2 control. Williams (1983) reported the active development of two high energy (400-800 keV) electron beam treatment processes and a high velocity gas discharge process as emerging alternatives to scrubbers. The pulse energized electron reactor (PEER) system utilizes electrons but represents a new departure in SO_2 control technology in so far as can be ascertained from available information. Operating on a gas stream which contained air, 1,666 ppm of SO_2, and water vapor, a bench-scale PEER system removed more than 90% of the SO_2 with a substantially smaller power requirement than is necessary for energetic electron beam treatment or the HV DC discharge process.

The high energy electron beam treatment processes are understood as means of producing oxidizing radicals (O^-, OH^-, and HO_2, etc.). The radicals react with SO_2 and form aerosols which can be collected by an electrostatic precipitator or a bag filter. The electron beam process promises advantages over scrubbers, but it requires multiple accelerators which are capital intensive and x-ray shielding is necessary (Bush, 1978). In the PEER system, a pulse streamer corona discharge produces the desired radicals. Since the formation energy of the radicals is only 5-15 eV, many of the electrons in the corona streamer have sufficient energy.

The use of DC and AC coronas for the removal of NO from flue gas has been previously reported by Tamaki (1979). These processes were power inefficient and the performance was poor. A possible explanation of the poor performance is the size of the ionization region for the DC coronas. A low power efficiency would result from a dissipation of power on ionic migration.

The need for power efficient enhancement of dose in practical applications of treatment by electrons has been discussed by Davis (1982). The development of the PEER device began with dose enhancement as an objective, but as results will show, the PEER system is self-contained and is most advantageously used a stand alone device (Mizuno, October, 1984).

The PEER chamber shown in Fig. 11 is a multi-purpose device which can operate as a self-contained PEER device, as a DC discharge treatment device, as an energetic electron beam reactor or with two or more of these operating modes in combination. The chamber is a rectangular plexiglass box with one end covered by a thin plastic film which serves as a window for energetic electron beam treatment of the entire chamber volume. The volume of the chamber is 9.2 ℓ. For a gas flow rate of 1.2 ℓ/min., the average electron beam treatment time is 7.6 minutes. Energetic electron beams were produced by a 3 MeV Van de Graaff accelerator operated at a beam energy of 1.2 MeV and the beam currents used were in the range of 0.5-10 μA. The radiation dose delivered to the gas is proportional to the beam current and to the actual energy loss of the beam in the gas of the chamber. The dose rate measured at the center of the reactor chamber (33 cm from the foil window) was 0.017 Mrad/min. when a beam current of 0.5 μA was used. A CTA film dosimeter was used for the dose measurements.

The has handling system is shown in Fig. 12. The model gas, a mixture of air, SO_2 and water vapor was introduced to the reactor chamber at room temperature (22°C). The SO_2 concentration was 1,666 ppm, the H_2O concentration was 2.5% by volume (100% RH), the flow rate was 1.2 ℓ/min. A pulsed fluorescent SO_2 analyzer measured the SO_2 concentration of the gas leaving the reactor chamber.

Pulsed energization was provided by the power supply shown in Fig. 13. The output of this supply is a voltage pulse with a peak voltage (V_p) range of 30-50 kV, with a width of approximately 200 ns, and a frequency of 60 Hz. A DC bias voltage (V_{dc}) of 0-50 kV can be applied.

The results of experiments in which the PEER chamber was used as a stand alone positive pulsed streamer corona device, a DC discharge chamber, an energetic electron treatment chamber or some combination of these modes are given in Table I and in Fig. 14. Experiment No. 9 of Table I shows the advantage of the PEER process over the other alternatives or combinations of processes. In this case the pulse voltage was +45 kV and the DC bias was +20 kV.

The two columns at the right give the delivered power for the operating mode and the electron beam power which would be required to achieve the same performance. In the case of experiment No. 9, the power efficiency advantage is greater than 3. In run No. 4, only positive pulsed energization was used and the power advantage is greater than 2 over that for energetic electron beam treatment.

Experiments No. 1 through No. 7 were performed with a rod and plate geometry, while experiments No. 8 through No. 11 are the result of a needle and plate geometry.

An advantage of the pulsed mode of operation is that because of the short duration of the pulses, a peak voltage can be delivered which is significantly larger than the DC breakdown voltage. The short pulse duration yields a large power efficiency since the ions do not move significantly during the pulse and negligible energy is wasted on ion migration. The pulses are of sufficient duration to energize electrons which produce the radicals.

The estimate of the exposure time to the positive pulsed streamer coronas is approximately 7 seconds. This is determined by the ratio of the volume occupied by the discharge to the volume of the chamber and the gas residence time of 7.5 min. The pulse discharge volume is assumed to be a hemisphere of a radius equal to the distance between the electrode and the plate. These experiments show that an exposure time of the order of 1 second can be readily achieved with high removal efficiency.

Figure 14 gives a plot of the comparisons given in Table I. The advantage of the PEER device is graphically shown by the low value of the penetration in experiment No. 9 for which the energetic electron beam current was 0, i.e., the accelerator was used.

In a separate experiment which is reported in a paper by Mizuno et al. at this conference, dust particles were introduced into the gas stream to better model a flue gas. When the collecting electrode was covered with a high resistivity layer, particle collection efficiency for pulsed streamer corona operation was better than that using DC voltage only, the conventional mode for electrostatic precipitator operation. In all of the comparisons of positive polarity versus negative polarity, the performance of positive pulse voltage was superior, an advantage qualitatively explained by the larger volume of discharge encompassed for the positive polarity as compared with the negative polarity.

CONCLUSIONS

(1) The energy of electrons used in the treatment of flue gas for the removal of noxious gas factions or particulate matter can be controlled in various energizing mechanisms and this control is crucial to optimum performance efficiency and power consupmtion efficiency.

(2) A bi-electrode low energy electron beam precharger has charged 1 and 3 μm diameter particles to charge values higher than those given by the theory for ionic charging by a factor of 5 for 1 μm. The increase in charge may be due to space charge enhancement of the electric field and/or by free electron diffusion charging.

(3) Pulsed positive streamer corona treatment provided by a pulse energized electron reactor (PEER) has removed more than 90% of the SO_2 from a test gas stream with a power efficiency greater than that for energetic electron beam treatment by a factor of at least 2 and with an acceptable exposure time.

(4) Preliminary experiments reported elsewhere at this conference suggest that both SO_2 and suspended dust particles can be removed in a modified PEER system which operates with pulsed voltage and a DC bias voltage.

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TABLE I

SO_2 REMOVAL RESULTS

No.	Treatment Combination	EB Current (μA)	V_p (kV)	V_{dc} (kV)	I_{total} (μA)	SO_2 Penetration (%)	Delivered Power (W)	Equivalent EB Power (W)
1	EB	1.0				92	0.07	
2	EB	5.0				46	0.34	
3	EB	10.0				22	0.68	
4	$+V_p$		+45		+5	30	0.23	0.58
5	$+V_p$+dc		+45	+20	+8	15	0.28	0.91
6	EB+V_p	5.0	+45		+5	25	0.57	0.65
7	EB+dc	5.0		+33	+115	36	4.14	0.49
8	+dc			+30	+26	78	0.78	0.13
9	$+V_p$+dc		+45	+20	+10	6	0.31	1.34
10	-dc			-50	-125	54	6.25	0.30
11	$-V_p$-dc		-45	-20	-4	45	0.07	0.38

No. 1-7 Rod to plate electrode
No. 8-11 Needle to plate electrode

F.S.U. ELECTRON BEAM PRECIPITATOR
TEST SYSTEM

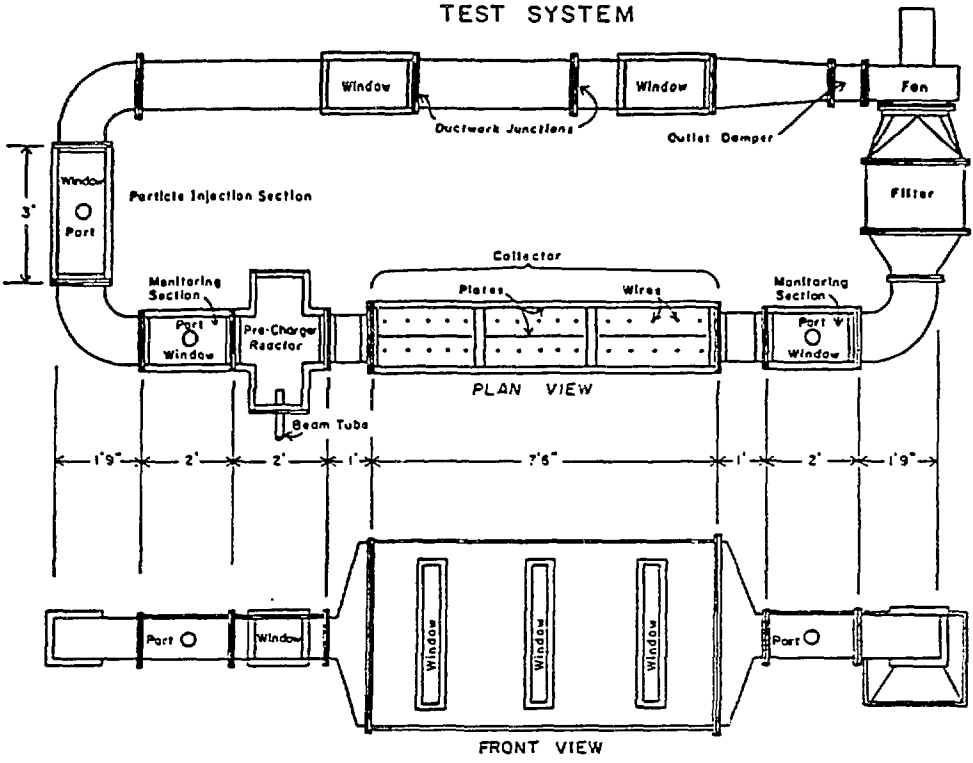


Figure 1

PRECHARGING MODULE (Top)

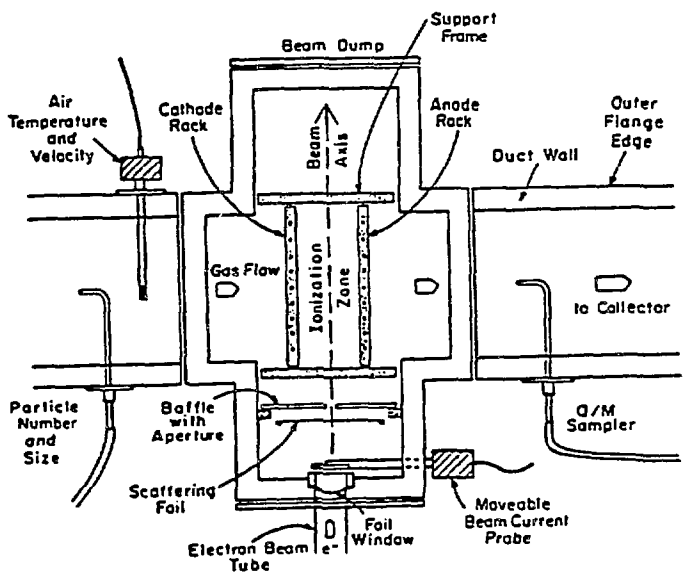


Figure 2

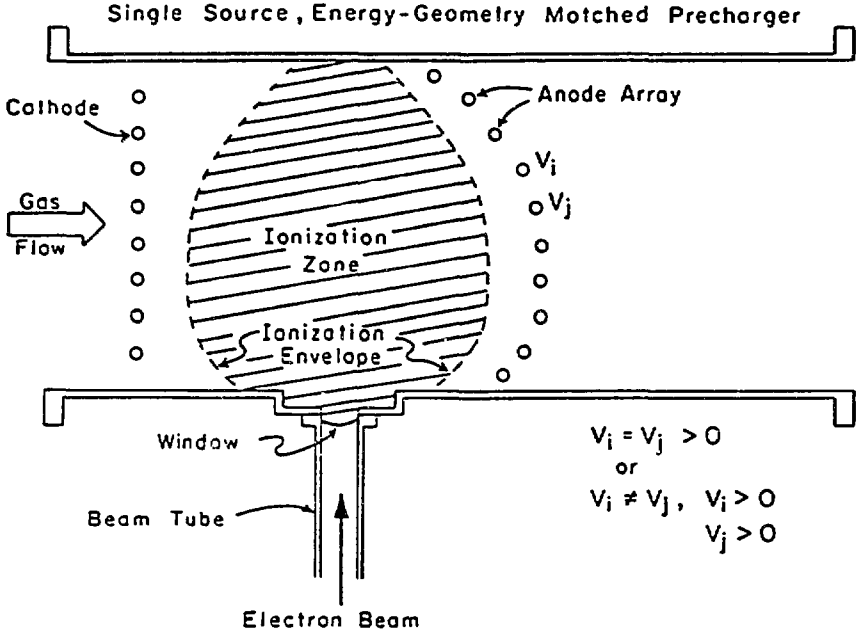


Figure 3

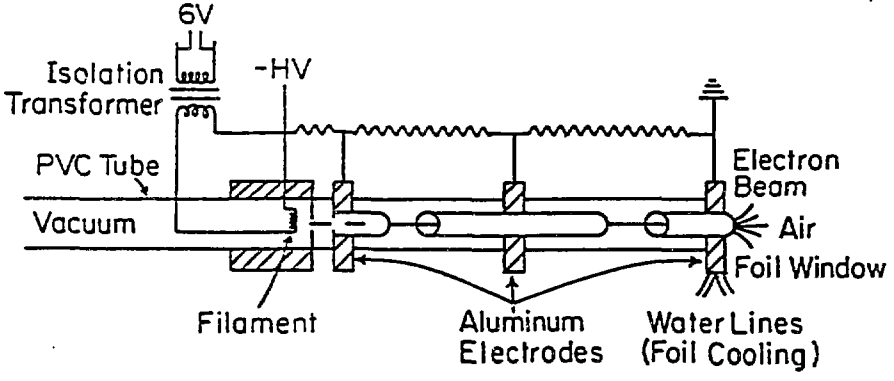


Figure 4

TRI-ELECTRODE E-BEAM PRECHARGER

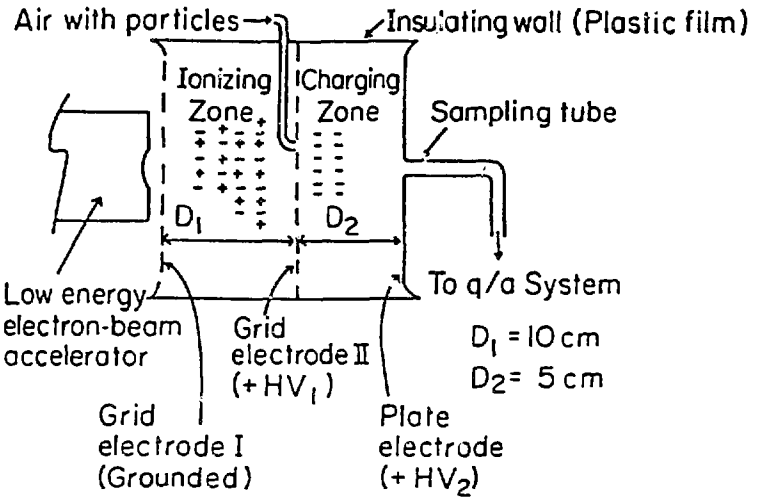


Figure 5

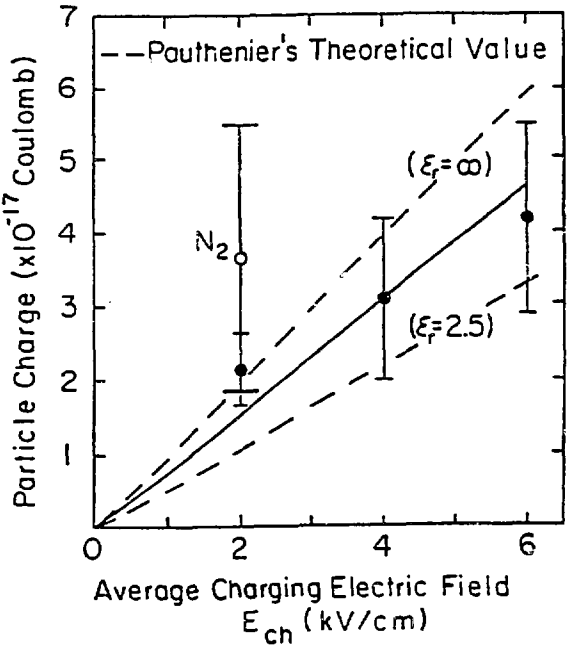


Figure 6

BI – ELECTRODE E-BEAM PRECHARGER

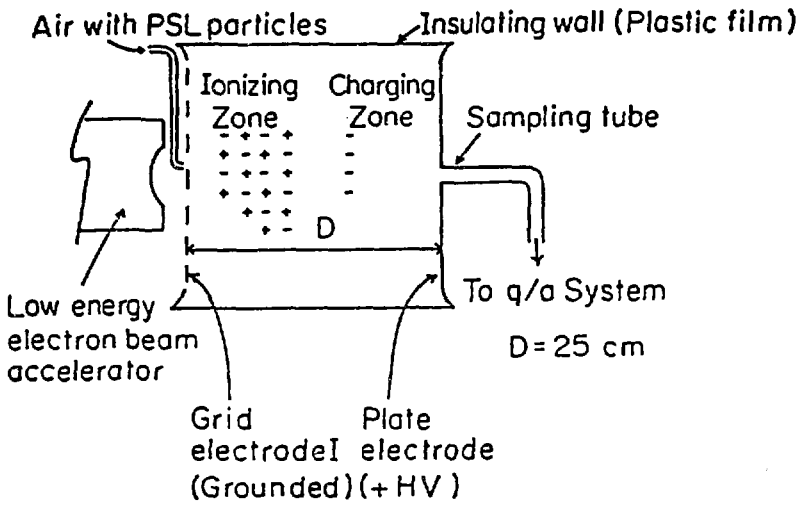


Figure 7

BI-ELECTRODE LOW ENERGY ELECTRON BEAM PRECHARGER
1.1 μM PSL PARTICLES

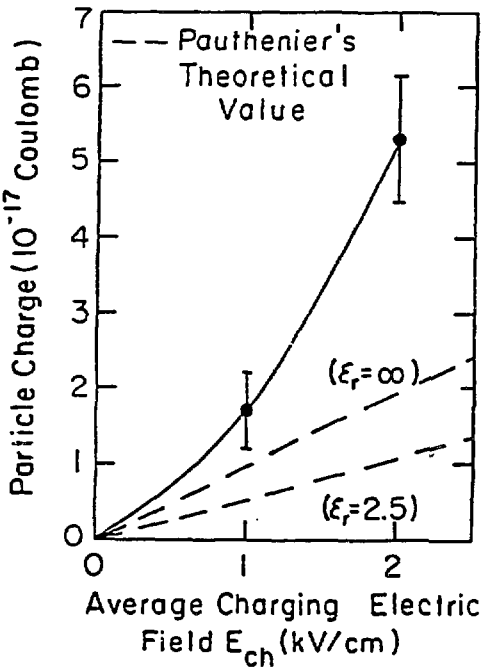


Figure 8

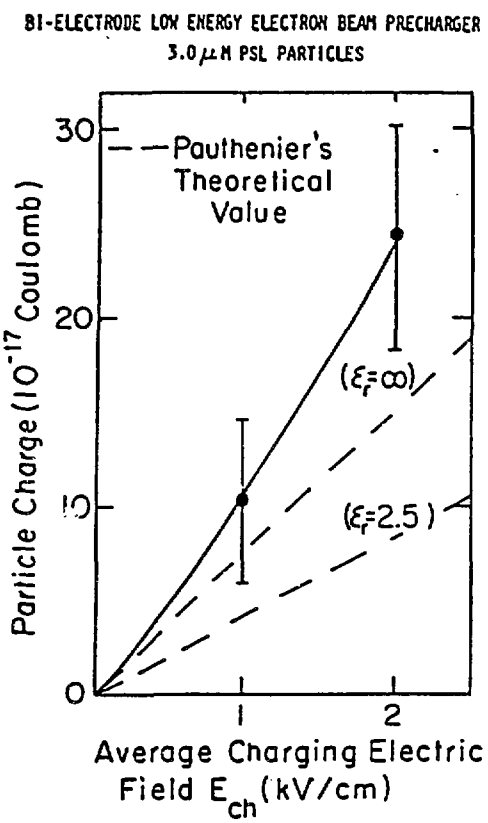
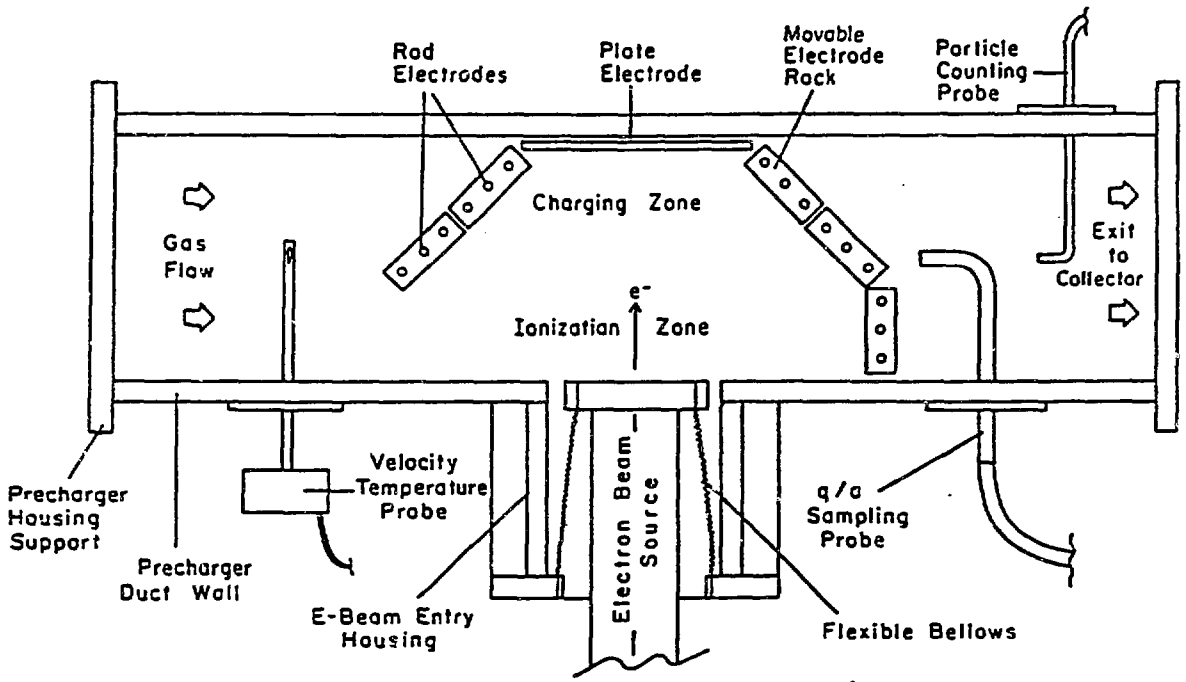

BI-ELECTRODE LOW ENERGY ELECTRON BEAM PRECHARGER
3.0 μM PSL PARTICLES
30
20
10
0
Particle Charge (10^{-17} Coulomb)
Pauthenier's Theoretical Value
($\varepsilon_r=\infty$)
($\varepsilon_r=2.5$)
1
2
Average Charging Electric Field E_{ch} (kV/cm)

Figure 9



A Precharger Housing (Top View, $\frac{1}{4}$ scale)

Figure 10

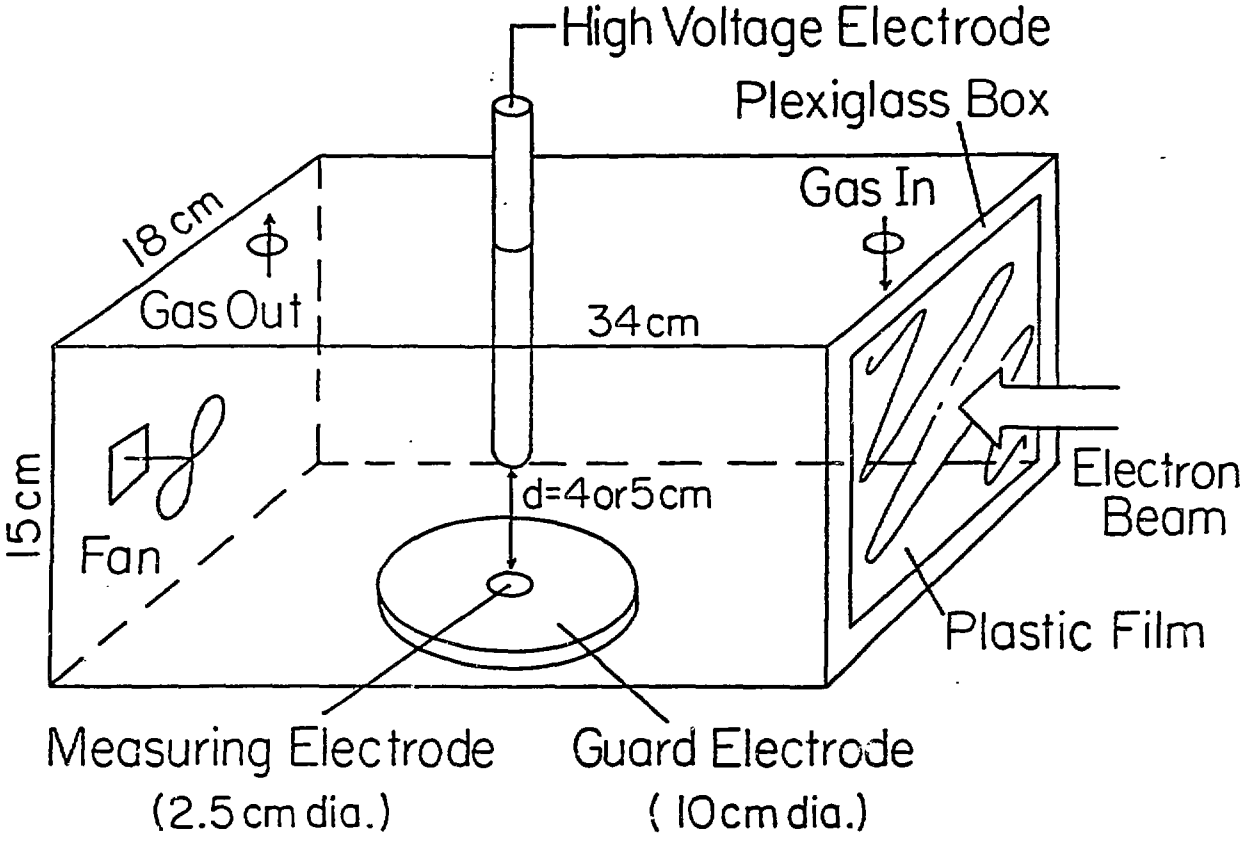

High Voltage Electrode
Plexiglass Box
Gas In
18 cm
Gas Out
34 cm
15 cm
Fan
d=4or5cm
Electron
Beam
Plastic Film
Measuring Electrode
(2.5 cm dia.)
Guard Electrode
(10cm dia.)

Figure 11

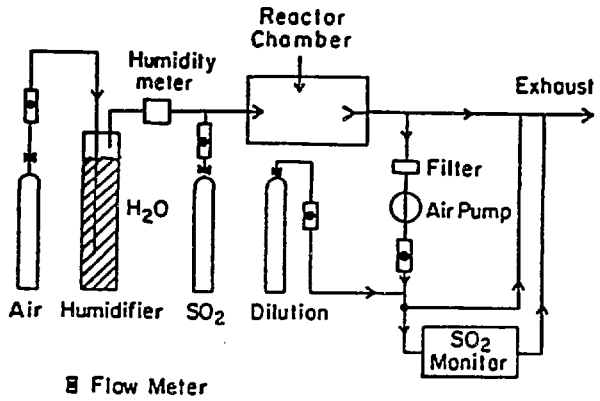


Figure 12

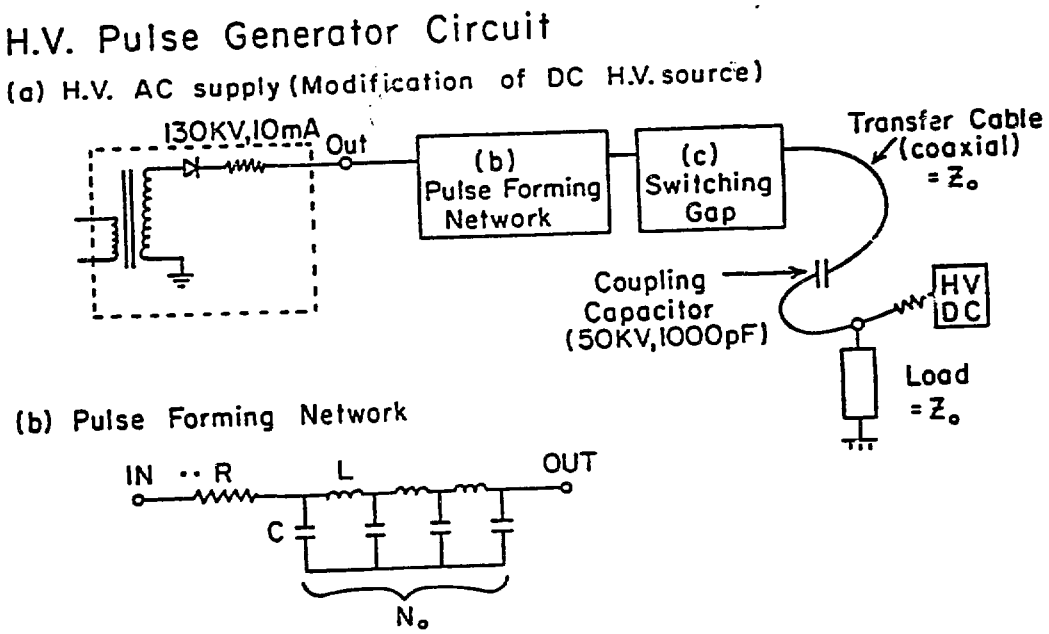


Figure 13

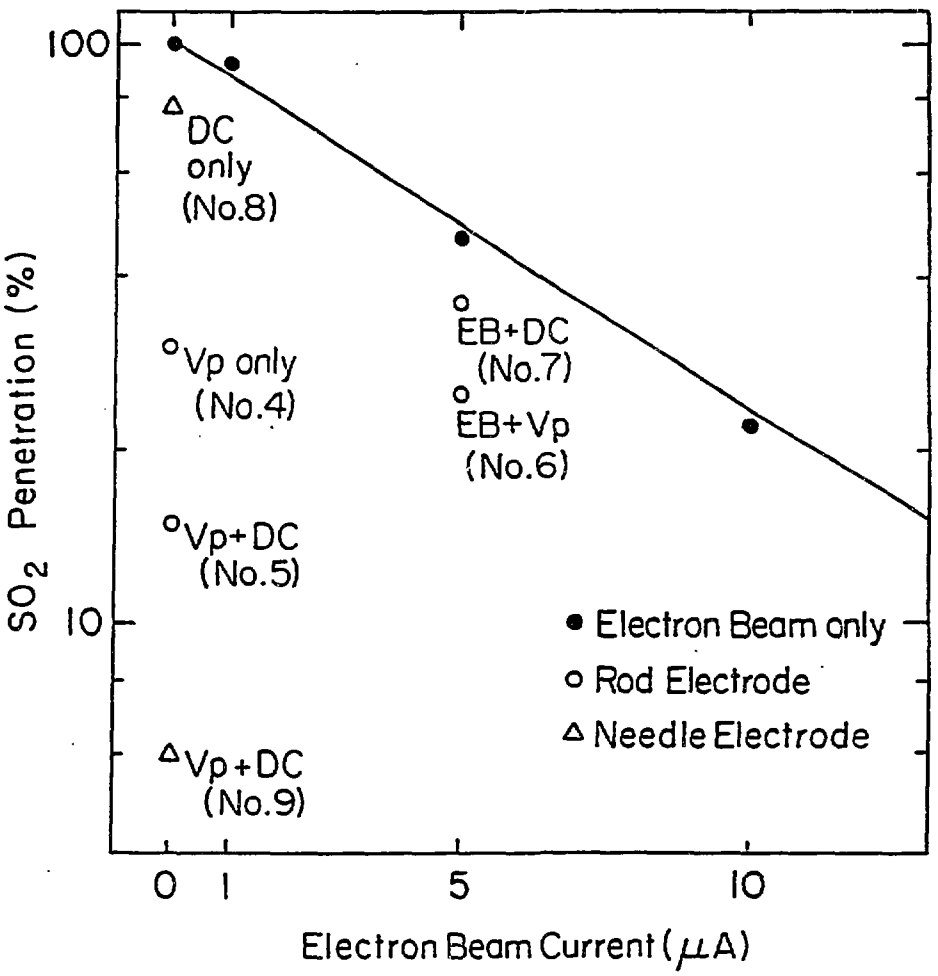


Figure 14

DISCLAIMER