A HIGH TEMPERATURE PULSED CORONA PLASMA REACTOR

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Abstract

Non-thermal plasma reactors have recently been used for the treatment of gaseous pollutants. High energy electrons (several eV) are produced in the plasma while the gas remains near ambient temperatures. Pollutant molecules are decomposed by highly reactive chemical radicals created through electron collisions. The focus of this work is the treatment of pollutants from the exhaust of electric arc incinerators. A pulsed corona reactor capable of operation at exhaust temperatures of hundreds of degrees C has been constructed. This design can be used as a conventional pulsed corona reactor (wire-metal tube geometry) and has the potential for use as a hybrid reactor which incorporates a wire-tube geometry with a ceramic dielectric barrier on the inside surface of the metal tube. Pulse widths of a few 10's of ns and risetimes of less than 10 ns have been obtained. Specifically, the reactor performance as a function of temperature is investigated. The preliminary results of the destruction of gaseous pollutants from this prototype are presented along with electrical and chemical efficiencies of the device.

Introduction

Non-thermal plasmas are gaining interest in a variety of applications. The ability to produce high-energy electrons in a neutral gas at atmospheric pressures while keeping the ambient temperature unchanged is very attractive for cleaning of gaseous effluents. The three most common methods of producing a non-thermal plasma are the irradiation of a gas with a high-energy electron beam, the use of a dielectric barrier discharge (also called a silent discharge), and the use of a pulsed corona discharge [1,2,3]. Both the dielectric barrier discharge and the pulsed corona discharge introduce the energy into the gas by the production of a transient electron avalanche discharge. The dielectric barrier discharge is a self extinguishing electron avalanche discharge (commonly referred to as a microdischarge) which is terminated by charge buildup on the dielectric. A pulsed corona reactor (PCR) uses a coaxial geometry, typically a thin wire of tens to hundreds of mils diameter is used as the center conductor with a metal tube outer conductor, and is driven by a high-voltage pulse generator. The pulse must be kept within a few hundred nanoseconds to avoid the transition of the non-thermal discharge to a thermal arc discharge. Similar to the dielectric barrier discharge, corona discharges streamers are created along the length of the reactor tube (estimated to be 10's per centimeter) and are terminated by the field inhomogeneity inherent in this geometry.

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14. ABSTRACT
Non-thermal plasma reactors have recently been used for the treatment of gaseous pollutants. High energy electrons (several eV) are produced in the plasma while the gas remains near ambient temperatures. Pollutant molecules are decomposed by highly reactive chemical radicals created through electron collisions. The focus of this work is the treatment of pollutants from the exhaust of electric arc incinerators. A pulsed corona reactor capable of operation at exhaust temperatures of hundreds of degrees C has been constructed. This design can be used as a conventional pulsed corona reactor (wire-metal tube geometry) and has the potential for use as a hybrid reactor which incorporates a wire-tube geometry with a ceramic dielectric barrier on the inside surface of the metal tube. Pulse widths of a few 1 O's of ns and risetimes ofless than 10 ns have been obtained. Specifically, the reactor performance as a function of temperature is investigated. The preliminary results of the destruction of gaseous pollutants from this prototype are presented along with electrical and chemical efficiencies of the device.

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The focus of this work is to develop a non-thermal plasma generator capable of operating at the high ambient temperatures (up to 500 C) produced at the exhaust of an arc melter solid waste treatment system being developed for the Buried Waste Integrated Demonstration. There are two main reasons for choosing a pulsed corona geometry over a dielectric barrier geometry for this application. The dielectric loss of most dielectrics increases at higher temperatures which could greatly decrease the overall efficiency of a dielectric barrier discharge and may lead to a thermal run away effect in the dielectric. Another area of concern is the small gap spacing of the dielectric barrier discharge which is easily blocked by solid particulates. A PCR discharge would be less susceptible to these problems. Arcing is a problem in conventional PCRs, however a dielectric barrier may be placed in the low field region at the outer conductor to inhibit any arcing that may occur. The dielectric loss in this configuration is low since it is positioned in a low field region and the frequencies are much higher than in the conventional dielectric barrier discharges.

Experimental Setup

The experimental apparatus is shown in Fig. 1, which illustrates the pulsed corona reactor, the electrical diagnostics, and the chemical diagnostics. The gas mixture flows continuously through the reactor and the contaminant concentration is monitored with the gas chromatograph at the output. The PCR is then be turned on and parameters such as repetition rate can be varied. A measure of the effectiveness of the reactor for the destruction of various contaminants is determined by combining the measurements of the deposited electrical energy and the pollutant concentration removed.

The PCR is constructed in a coaxial geometry to reduce electromagnetic interference and to maintain a fast pulse risetime. The PCR is driven by a self-breaking high pressure hydrogen spark gap. A constant-voltage, high-voltage power supply is used to charge the storage capacitance through a high voltage resistor. The repetition rate is set by the voltage level, the value of the charging resistor R_c, and the value of the storage capacitance. The energy stored in the RG-217 cable capacitance, either 80 pf or 235 pF, is resonantly transferred to the reactor capacitance, approximately 20 pF, when the spark gap closes. The energy decay of the storage capacitance is determined by monitoring the voltage, V_c, at the storage capacitance using a Tektronix high-voltage probe, model P6015A. The output pulse is monitored using a fast capacitor divider high-voltage probe, V_{PCR} (sensitivity 3500V/V), and a fast current viewing resistor current probe, I_{PCR} (sensitivity 35A/V). These probes are built into the coaxial housing between the spark gap and the reactor. Each probe is capable of measuring down to a 1 ns risetime. The decay time of the voltage probe is 2 μs giving a window of accuracy of the probe of 200 ns. The energy per pulse is obtained by multiplying the voltage and current pulses and integrating over the pulse width. The measurement of the energy delivered from the storage capacitance and the energy delivered to the reactor is used to monitor the operation of the PCR and provides an accurate measure of the energy delivered to the gas flow.

A view of the reactor tube is also shown in Fig. 1. We have used two reactor configurations 1) a standard pulsed corona geometry (no dielectric barrier) utilizing a coaxial stainless steel wire and tube and 2) a hybrid PCR geometry utilizing an alumina tube placed on the inside of the outer conductor as the dielectric barrier. The reactor is two feet long with a 20 mil diameter wire and a 1 inch diameter outer conductor. The hybrid reactor can be operated in the

same manner as the conventional PCR with the additional requirement of removing the charge from the dielectric barrier between pulses. The hybrid PCR configuration is different from the typical dielectric barrier discharge since the streamers are initiated at the field enhancement region around the center conductor, as with the standard pulsed corona configuration.

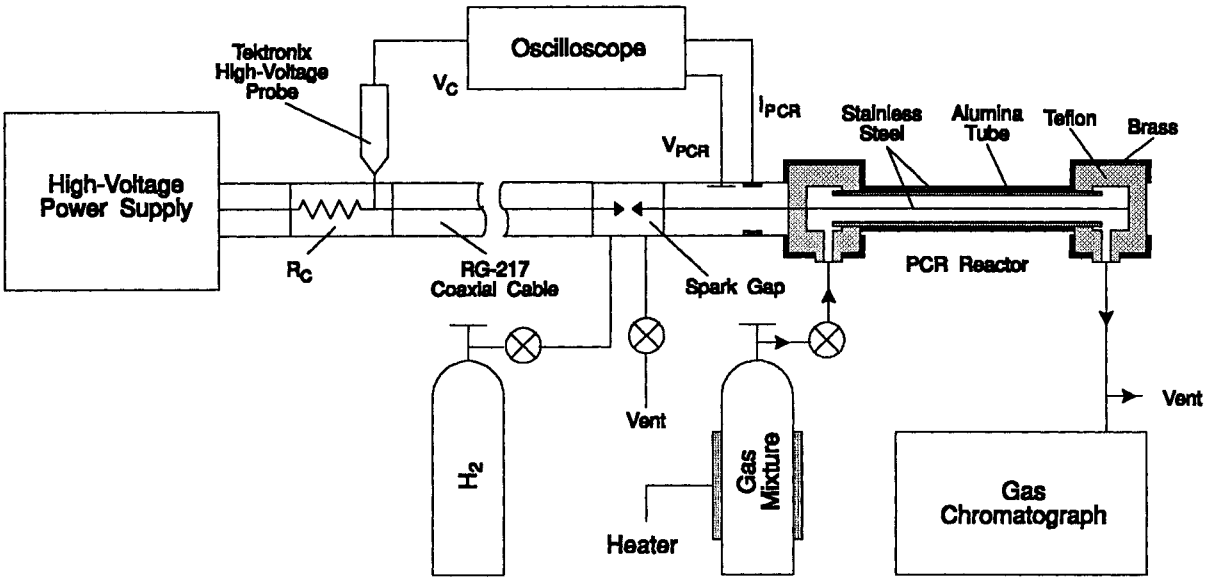

Figure 1. The experimental arrangement which includes the pulsed corona reactor, the electrical diagnostics, the gas flow apparatus, and the gas chromatograph.

A Perkin Elmer, model Sigma 2000, gas chromatograph is used to obtain destruction and removal efficiency (DRE) measurements for the PCR. The gas mixture, usually a few hundred ppm of pollutant in air, is mixed using a liquid nitrogen gas mixing station and placed in a heated gas cylinder. The gas mixture is continuously flowed through the PCR and the discharge is turned on and off and the discharge parameters are varied. The gas chromatograph is used to monitor the concentration of the pollutant.

Experimental Results

To effectively evaluate the performance of the reactors, the amount of energy deposited in the gas flow necessary to remove a fixed percentage of pollutant is measured. This is achieved by combining the measurement of energy delivered to the gas flow from the electrical diagnostics and the corresponding chemical destruction measurements from the gas chromatograph.

The current and voltage characteristics were measured as described above. Figure 2 shows a comparison between the current and voltage pulses obtained by driving the standard PCR configuration with an 80 pF and a 235 pF storage capacitance. The pulse width using the 80 pF capacitance is approximately 25 ns and the pulse width using the 235 pF storage capacitance is approximately 50 ns. The energy per pulse was approximately 30 mJ for the 80 pF storage capacitance and was 50 mJ for 235 pF. The risetime of the system is less than 10 ns for all PCR configurations. For the standard PCR configuration, the arcing threshold was different for the

two capacitance values. The arcing threshold is dependent on several parameters which include flow rate, peak voltage, and repetition rate. Therefore a convenient parameter for comparison is the energy density, E, that can be delivered to the gas before arcing occurs in the reactor. The values of E at the arcing threshold for the two storage capacitances were 1550 J/l for 80 pF at 350 Hz, 0.35 slpm, and a peak voltage of 27 kV and 800 J/l for 235 pF at 90 Hz, 0.3 slpm, and a peak voltage of 28 kV.

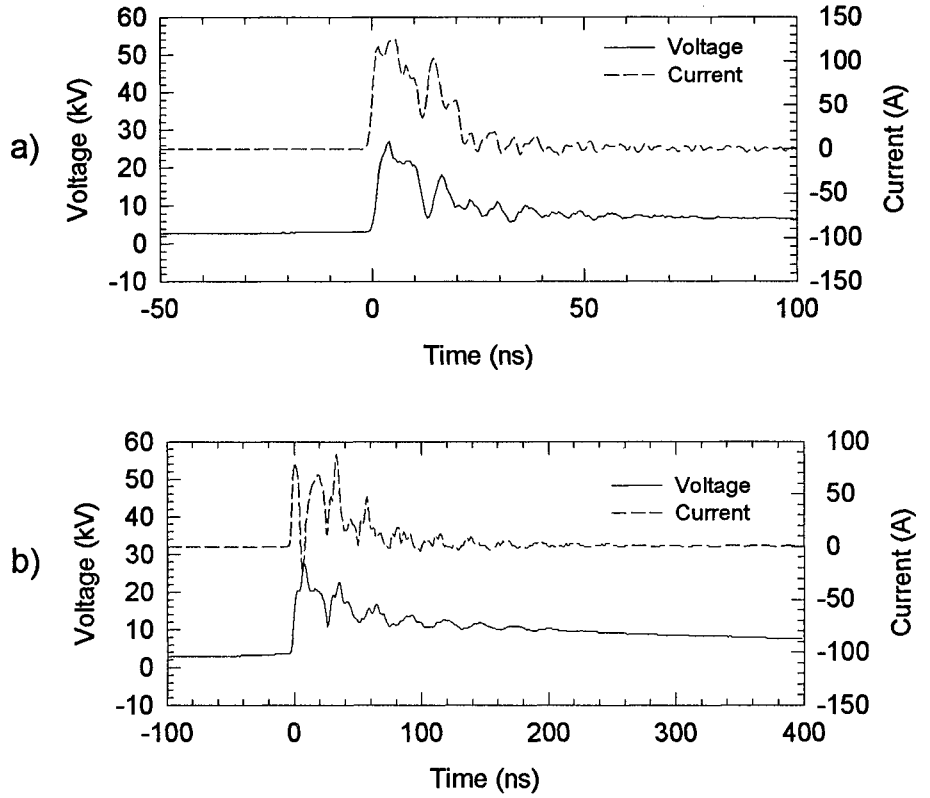

Figure 2. The current and voltage versus time for the standard pulsed corona reactor for a) an 80 pF storage capacitance and b) a 235 pF storage capacitance.

The pulse waveforms for the hybrid PCR configuration are similar to those shown in Fig. 2 for the standard PCR. However, using 80 pF as the storage capacitance, the pulse width for the hybrid configuration is approximately 20 ns. The peak voltage for the hybrid configuration was 22 kV and the energy per pulse was 20 mJ. The operation of the hybrid PCR reactor is markedly different from the conventional PCR reactor in that the arcing is effectively eliminated by the barrier under similar operating conditions. The shortening of the pulse is probably due to the extinguishing of the streamers by the dielectric barrier sooner than in the standard PCR. The end result is that the hybrid PCR can be run at much higher repetition rates. The present reactor has been operated to 800 Hz and is capable of much higher rep-rates, limited by the recovery characteristics of the hydrogen spark gap[4].

The standard PCR operation was investigated to 200 C. A slight increase in the arcing threshold was observed along with a decrease in the residual voltage left on the reactor after the pulse. The residual voltage dropped from 10 kV at room temperature to less than 1 kV at 200 C.

With the pulse voltage remaining the same, the E/N is increased due a decrease in the neutral number density at higher temperatures at atmospheric pressure. This may lead to increased streamer activity as well as an increase in the density of streamers per unit length in the reactor.

In preliminary experiments the destruction and removal efficiencies of trichloroethane (TCA) and methylene chloride have been measured. Figure 3-a shows the energy density versus DRE for TCA removal, 200 ppm initial concentration, in dry air using two different storage capacitors driving the conventional PCR reactor. As was discussed above, the maximum energy density with the 235 pF capacitor was 800 J/l compared to 1550 J/l using the 80 pF capacitor. Much higher destruction levels can be obtained for the smaller storage capacitor since the arcing threshold is increased. However the energy density versus DRE follows the same trend for the two storage capacitor values. In Figure 3-b the energy density versus DRE for three experimental conditions are shown. A direct comparison of the destruction of 200 ppm TCA in dry air between the conventional PCR reactor and the hybrid PCR reactor is seen. The hybrid reactor is slightly less efficient than the conventional PCR at the higher destruction rates. This may be due to a lower peak voltage, 22 kV, used during the operation of the hybrid reactor compared to 27 kV for the standard PCR configuration. However, overall the DRE for the two reactor configurations are very similar. A few data points were obtained for 200 ppm methylene chloride in dry air for comparison using the hybrid PCR configuration. Methylene chloride requires more energy to obtain the same amount of destruction as with TCA in dry air.

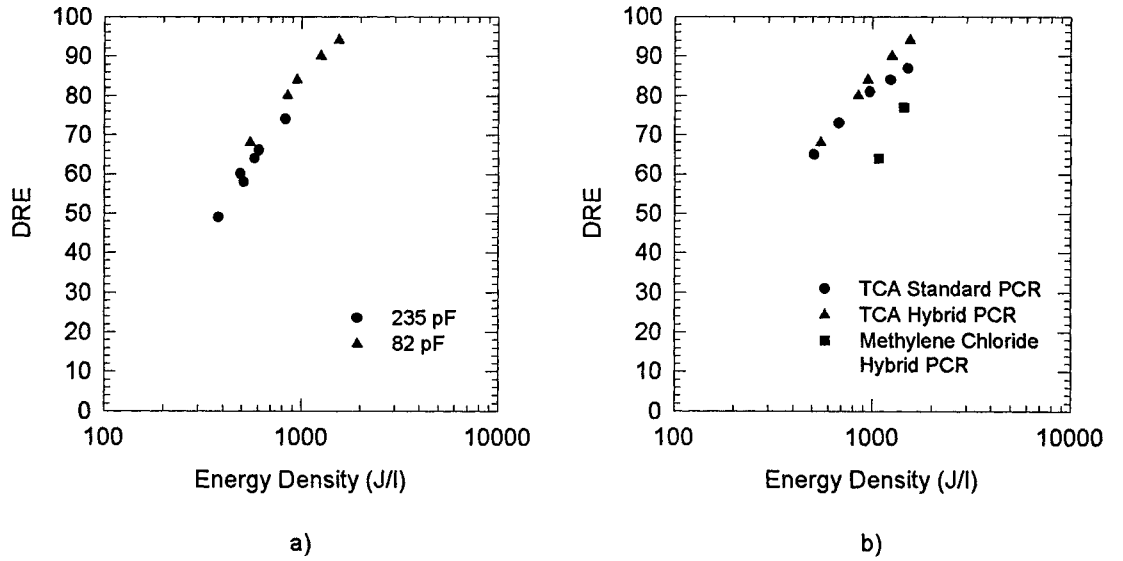

Figure 3. The energy density versus DRE for a) a comparison between the conventional PCR driven by two different storage capacitances and b) a comparison between the conventional and hybrid PCRs for TCA and methylene chloride in dry air.

In many cases, the removal of contaminant X can be described by an exponential dependence,

$$[X]=[X]_o\, 10^{-E/\alpha} \quad \text{or} \quad [X]=[X]_o \exp^{-E/\beta},$$

where [X] is the final concentration, $[X]_o$ is the initial concentration, E is the energy density delivered to the gas, α is the amount of energy necessary to obtain 90% removal and β is the amount of energy necessary to obtain 63% removal. The α and β values for the data acquired above are shown in Table I. The α and β values for TCA in dry air using three different PCR configurations are very close. It is also seen as stated before that a significantly higher energy is required for 90% destruction of methylene chloride compared to TCA in dry air. Only a preliminary DRE measurement was taken for 200 ppm TCA in dry air at 175 C and there is an indication that the α and β factors decrease with increasing temperature.

Table I. A comparison of α and β values for different PCR configurations and gas mixtures.

Configuration	α (J/l)	β (J/l)
PCR 235 pF, 200 ppm TCA	1300	560
PCR 82 pF, 200 ppm TCA	1200	520
Hybrid PCR 80 pF, 200 ppm TCA	1380	600
Hybrid PCR 80 pF, 200 ppm Methylene Chloride	2200	950

Conclusions

The construction of a standard pulsed corona reactor and a hybrid pulsed corona reactor has been completed for high temperature, flue gas treatment. Energy density figures of merit, α and β, required for a fixed percent reduction of the pollutants, 90% and 63% respectively, have been presented. The operation of the hybrid reactor is nearly the same as that for the standard PCR with the added advantage of higher repetition rate operation. Low temperature operation and chemical destruction measurements indicate that the best configuration is to use a hybrid reactor with a small storage capacitance at high repetition rates. Operation of a standard PCR reactor has been demonstrated to 200 C and preliminary chemical destruction measurements indicate increased destruction of TCA at elevated temperatures.

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