CHEMICAL DESTRUCTION USING A PULSED CORONA REACTOR

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Abstract: Chemical destruction measurements for a Blumlein driven pulsed corona reactor are presented along with observed current and voltage waveforms. The input gas flow consists of 0.2 to 1.5 SLPM room temperature air with a 200 ppm toluene impurity. The PCR is operated with an applied voltage of between 12 to 30 kV at 5 to 50 Hz repetition rate. The dependence of the PCR chemical destruction on rep-rate, applied voltage and flow are reported. Apparatus to observe the relation between the emission spectra and the chemical destruction is discussed.

INTRODUCTION

Many military and industrial processes produce hazardous gas by-products and environmental responsibility requires the implementation of abatement technologies. Furthermore, failure to satisfy international, federal, state and local government air pollution regulations can generate costly fines and impose serious operational delays. Nonthermal plasma discharge devices, such as the Pulsed Corona Reactor (PCR), promise energy efficient chemical destruction of unwanted molecules in contaminated gas streams[1]. On-going work with a wire-in-tube PCR has demonstrated excellent abatement performance[2,3] but some applications require higher chemical destruction efficiencies with less energy input. The numerous important physical processes (streamer shape / electron energy distribution, nonthermal reaction chemistries, . . .) involved in non-thermal gas abatement make theoretical prediction of the PCR difficult. We report here initial observations of a versatile, single tube PCR designed to determine the influence of operating parameters on chemical destruction. The PCR is designed to allow spectroscopic observations since the self luminous nature of the streamer discharges occurring in the PCR may provide a tool in the search for conditions and processes which lead to improved device operation.

REACTOR DESIGN AND OPERATION

An optically accessible pulsed corona reactor tube has been designed and built (see Fig. 1). The 20 mil diameter stainless steel wire is held in place by a stainless steel nub / o-ring assembly compressed against quartz end windows by wire tension. The 0.8 inch ID outer perforated stainless steel cylinder allows side-on optical access.

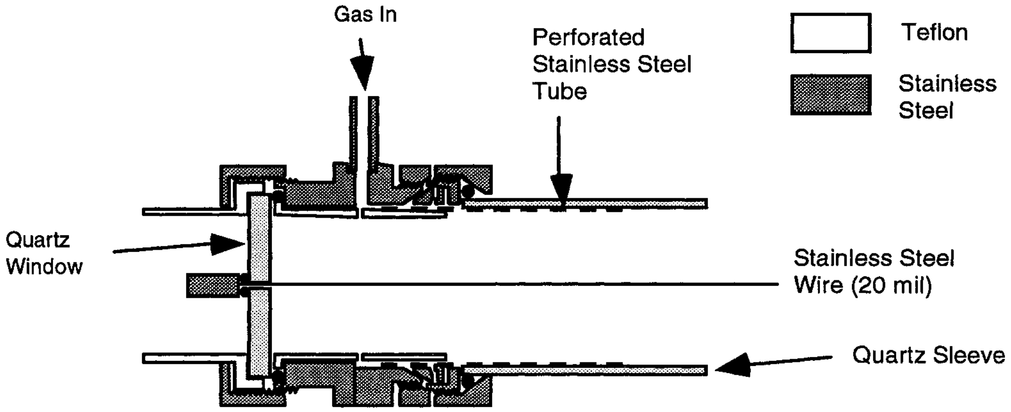

Fig. 1. Cross sectional view of one end of the PCR reactor assembly.

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14. ABSTRACT

Chemical destruction measurements for a Blumlein driven pulsed corona reactor are presented along with observed current and voltage waveforms. The input gas flow consists of 0.2 to 1.5 SLPM room temperature air with a 200 ppm toluene impurity. The PCR is operated with an applied voltage of between 12 to 30 kV at 5 to 50 Hz repetition rate. The dependence of the PCR chemical destruction on rep-rate, applied voltage and flow are reported. Apparatus to observe the relation between the emission spectra and the chemical destruction is discussed.

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Moderate risetime (< 20 ns) voltage pulses are applied between the wire and the outer tube using the 100 Ω impedance Blumlein driving circuit depicted in Fig. 2. The two 55 foot long RG8 cables comprising the Blumlein are charged through a 500 kΩ resistor and are discharged into a 100 Ω load resistor via a triggered spark gap switch pressurized with air. The PCR is placed in parallel across the load resistor with voltage between the wire and tube measured with a Tektronix voltage probe and with discharge current measured using a 10 ns risetime Pearson probe. This experimental design allows for variation in pulse width, applied voltage (at least 12 to 30 kV, dual polarity), rep-rate (single shot to 50 Hz) and risetime.

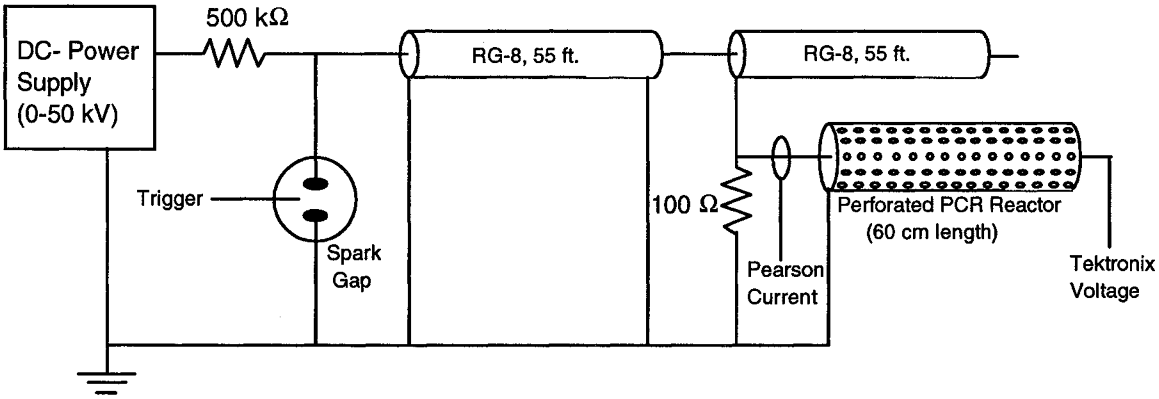

Fig. 2. Blumlein circuit used to create the PCR discharges.

Relatively square voltage pulses can be applied to the PCR since the discharge impedance stays above the 100 Ω load resistance (see Fig. 3) ; however, a small reflection pulse can be seen on the voltage waveform due to the finite PCR resistance.

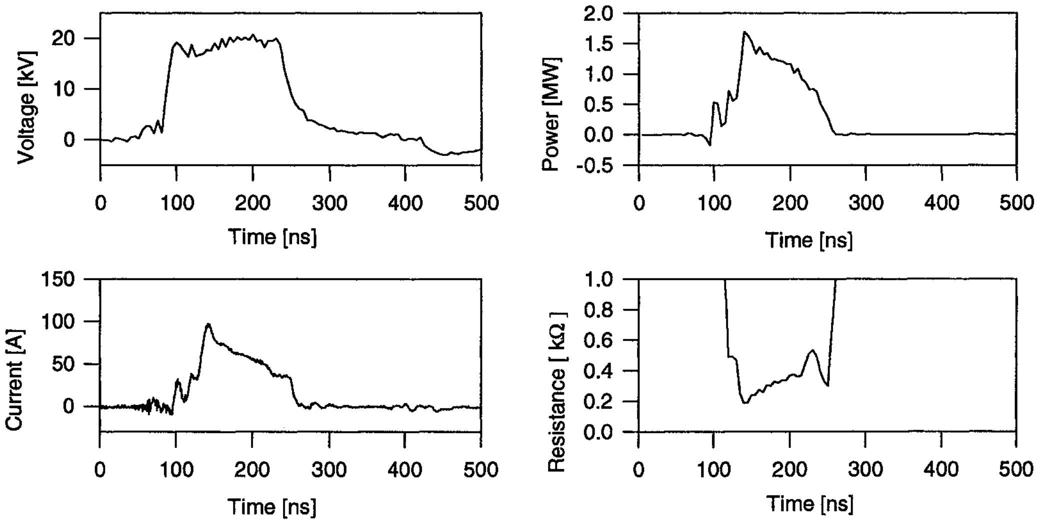

Fig. 3. Voltage, current, power and resistance waveforms for the Blumlein driven PCR.

Stray inductance in the switch portion of the driving circuit limits the applied voltage risetime to approximately 17 ns. The current shows a jagged rise to peak amplitude, the jaggedness due partially to displacement current. The current gradually declines and then abruptly drops at an applied voltage threshold of 10 kV even though the voltage across the wire-tube gap stays finite for > 100 ns.

CHEMICAL ANALYSIS

Contaminated gas streams were generated with the gas blending system shown in Fig. 4 where chemical destruction efficiency was measured using a gas chromatograph (GC).

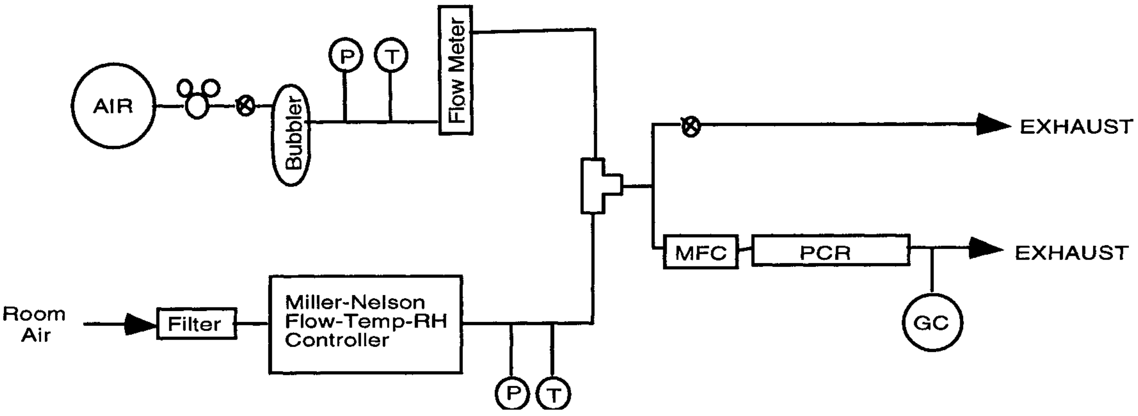

Fig. 4. Chemical delivery and analysis system.

A typical gas chromatogram showing toluene concentration versus experiment time for various voltages is shown in Fig. 5. With a few exceptions, the reactor came to equilibrium quickly allowing for good reproducibility and low uncertainty in the destruction measurements. The contaminated gas stream was generated by bubbling air through a container filled with toluene (the bubbler in Fig. 4). The toluene-air mixture was blended with the main carrier gas flow from the Miller-Nelson flow-temperature-relative humidity controller and introduced into the PCR through a sensitive mass flow controller (MFC) with (0.2 to 1.5 SLPM flow rates) Although an air-toluene mix was used in the work reported here; other carrier gases (He, N_2 . . .) and other liquid pollutants can be used with this system. Gas phase pollutants can also be mixed into the main gas flow.

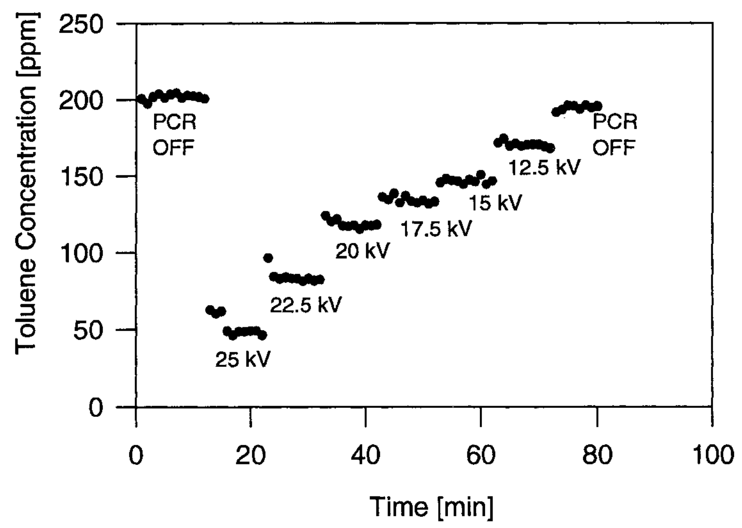

Fig. 5. Sample chromatogram showing toluene concentration dependence on applied voltage.

The analyte concentration from the bubbler (C_A in ppmv) can be calculated according to equation (1):

$$C_A = \frac{\chi_A \; P_A \; Q_A \times 10^6}{P_D \, Q_D + P_A \, Q_A} \qquad (1)$$

where $\chi_A = P_{sat} / P_B$ = mole fraction of analyte calculated from the saturation vapor pressure of analyte (P_{sat}, calculated from Antoine's equation using tabulated constants [4] and the temperature of the liquid analyte) and the total pressure in the bubbler (P_B), P_A = contaminant gas pressure, Q_A = volumetric flow rate of contaminant gas stream, P_D = main carrier gas pressure, and Q_D = volumetric flow rate of main carrier gas. Toluene chemical destruction efficiencies (DE) were calculated from Eq. (2):

$$\mathrm{DE} = \left[1 - \frac{C_{A\ ON}}{C_{A\ OFF}}\right] \times 100\% \qquad (2)$$

where $C_{A\ ON}$ and $C_{A\ OFF}$ are the concentrations of analyte measured using a gas chromatograph during the PCR on and off conditions, respectively. The gas chromatograph was calibrated daily with a custom standard cylinder containing 200 ppm (mole) of toluene in air. Taking into account the variations in the analyte supply concentration (Eq. 2) and uncertainties in the GC calibration gives a detection uncertainty for toluene on the order of 2 ppmv (S/N = 2).

RESULTS

Toluene destruction efficiency was measured while varying the applied voltage amplitude, the Blumlein rep-rate and the gas flow through the PCR all for 17% RH, room temperature air carrier gas with ~ 200 ppm toluene impurity. Figure 6 shows the toluene destruction efficiency versus the specific energy (average power deposited into the reactor per unit gas flow rate, Joules / Liter) for variations in applied voltage, rep-rate and flow. The specific energy was calculated by integrating the measured power into the discharge (voltage-current product), multiplying by the rep-rate and dividing by the PCR flow rate. The destruction efficiency and the Joules per Liter calculation each have an uncertainty of less than 10%.

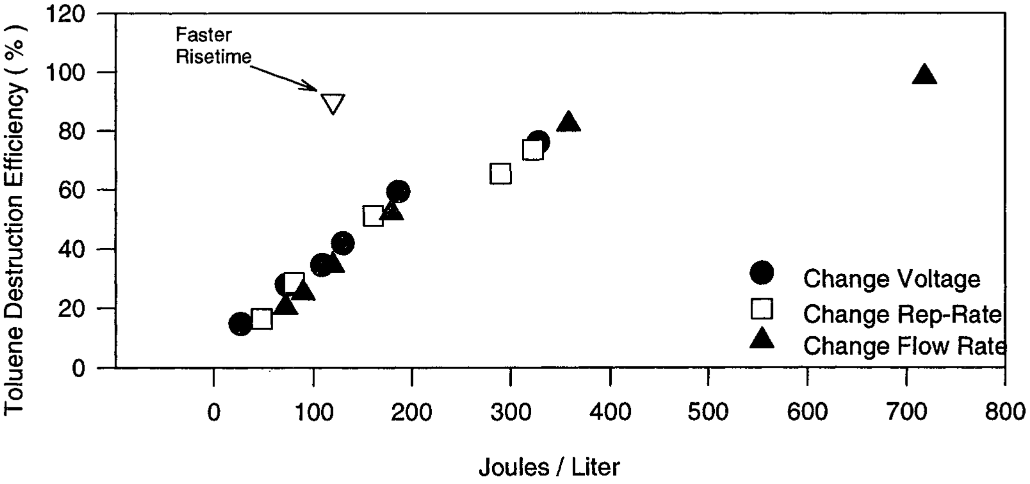

Fig. 6. Destruction efficiency of 200 ppm toluene mixed into 17% RH, room temperature air for various applied voltages, rep-rates and flow rates.

All the data tends to line up on a single, simple curve implying that destruction efficiency is dependent only on the Joules per Liter deposited into the reacting gas. However, the single data point labeled 'Faster Risetime' comes from measurements obtained with a 10 tube pulsed corona reactor device which had 6 ns voltage risetimes, 100's of Hz repetition rates, 100 ns current pulsewidth and 330 ppm toluene impurity. The better destruction measured with the 10 tube PCR implies that factors other than the specific energy are significantly influencing destruction - the voltage risetime, rep-rate or pulsewidth.

DISCUSSION

A hazardous molecule is stable because energy must be added to break the molecule apart. Since power must be supplied to cause chemical destruction, an unremovable cost of operating any abatement device is associated with the device power consumption. Therefore, since a given abatement application will typically have a required flow and desired destruction efficiency, the specific energy curve (as in Fig. 6.) for a particular device will dictate how much energy (and hence cost) is required for the application.[5] Clearly, the steeper the specific energy curve and the closer it is to the destruction axis, the cheaper a given cleanup operation. Comparison of specific energy curves aids in the choice of a cost effective abatement technology. Therefore, one goal in advancing pollution abatement state-of-the-art is to find some fundamental limits for a particular technology which will indicate just how good the best specific energy curve can be. For example, if the single data point on Fig. 6 is due to risetime, then efforts can be focused on making a faster risetime device or in postulating a minimum possible risetime.

On a microscopic basis, the fundamental improvement displayed by the 'Faster Risetime' data point on Fig. 6 indicates that perhaps the high energy electrons in the streamer tip are having a significant influence on the chemical destruction. How the energy is put into the gas appears to be important.

Future work involves extending our measurements to determine the dependence of chemical destruction efficiency on current pulse width and voltage risetime. Furthermore, in an effort to further understand the connection between streamer properties and chemical destruction, a Princeton Instruments intensified CCD array mounted to a 2/3 meter Czerny-Turner spectrometer will be used to acquire emission spectra from the PCR. Light will be collected end-on from the PCR and emission from the whole discharge volume will be sampled . The CCD will be operated so as to take one time integrated spectrum for each of 100 successive shots allowing fast accumulation of average spectra. The CCD can also be gated to less than 5 ns.

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