

# Corona Destruction: An Innovative Control Technology for VOCs and Air Toxics

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This paper discusses the work and results to date leading to the demonstration of the corona destruction process at pilot scale. The research effort in corona destruction of volatile organic compounds (VOCs) and air toxics has shown significant promise for providing a valuable contribution to critical U.S. Environmental Protection Agency and national goals of reducing the health effects associated with exposures to hazardous air pollutants. The corona destruction technology could be especially useful in future years in helping industry meet the residual risk requirements of the Clean Air Act Amendments of 1990.

Since 1988, EPA has conducted research in the area of corona destruction of VOCs and air toxics. EPA's interest in corona destruction of molecular species started with modeling of a point-plane reactor for destroying toxic organic compounds. EPA's goal is to develop a technology capable of controlling low concentration streams at low capital and operating costs. The purpose of this work is to develop an industrial scale corona reactor capable of efficiently and cost-effectively destroying VOCs and air toxics at ambient temperature and pressure. Results show that corona destruction is a promising control technology for many VOC-contaminated air streams, especially at low concentrations. Cost comparisons are presented for corona destruction and conventional control devices, carbon adsorption, catalytic incineration and thermal incineration.

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Since 1988, EPA's Air and Energy Engineering Research Laboratory has conducted research in the area of corona destruction of VOCs and air toxics. EPA's interest in corona destruction of molecular species started with modeling of a point-plane reactor for destroying toxic organic compounds.<sup>1</sup> The emerging concern for excessive concentrations of ambient ozone, for which many VOCs are precursors,

the need to develop technology to control low concentration streams, and the economic advantages of ambient temperature operation provided impetus for the work on high intensity corona reactor devices.

The purpose of this work is to develop an industrial scale corona reactor capable of efficiently and cost-effectively destroying VOCs and air toxics at ambient temperature and pressure.

The work and results to date leading to the demonstration of the corona destruction process at pilot scale are presented in this paper.

## Background

The initial work at EPA involved investigating the viability of corona destruction as an effective device for VOCs. The initial tests were run with toluene and were very successful. The next phase of the research program involved developing the destruction mechanism and prediction theory for destruction of other hydrocarbons. Even though corona destruction was able to destroy toluene, an understanding of the reaction mechanisms, both chemically and electrically, is necessary before larger systems can be successfully designed. Thus, a series of experiments was conducted using 10 compounds, and the destruction efficiency and ionization potential of each compound was examined to determine the possible existence of a statistical correlation. The compounds tested were benzene, cyclohexane, ethanol, hexane, hexene, methane, methylene chloride, methyl ethyl ketone, styrene and toluene. Preliminary results indicate that a relationship exists between ionization potential and ease of destruction for VOCs, the lower the ionization potential, the easier the compounds are destroyed.

Corona destruction has several advantages over conventional control devices as shown in Table I. The compounds that we have tested have been oxidized to carbon dioxide, carbon monoxide, and water, plus, in the case of chlorinated compounds, chlorine and hydrochloric acid. The exhaust streams have been analyzed thoroughly with a gas chromatograph/flame ionization detector (GC/FID) and gas chromatograph/mass spectroscopy (GC/MS), and no intermediate compounds have been found. Since the contami-



**Table I.** Benefits of corona destruction over conventional technologies.

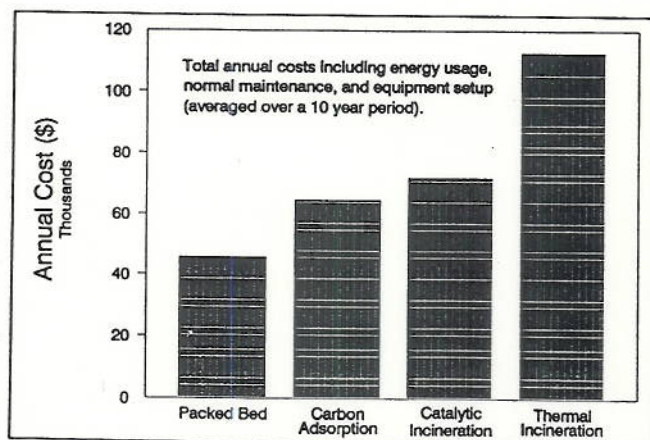
- Performs effectively and economically at very low concentrations
- Operates at ambient temperature
- Eliminates disposal or treatment problems associated with carbon adsorption
- Eliminates sensitivity to poisoning by sulfur or halogen containing compounds
- Requires no auxiliary fuel
- Requires low maintenance

nants are destroyed, the problem of disposing of collected toxins is avoided.

Two corona destruction processes have been evaluated for their potential in destroying VOCs and air toxics. One of the corona destruction processes uses high dielectric barium titanate pellets in a packed-bed reactor across which a high voltage alternating current (AC) is applied. The micro-electric fields developed in the interstitial spaces between the pellets form a multiplicity of corona sites which generate electrons. These electrons initiate the reactions that lead to destruction of the challenge gas species. The second process consists of a wire-in-tube reactor which is energized by high voltage nanosecond pulses. These techniques have the potential of generating very energetic electrons without wasting power by accelerating ions.

The corona processes operate at ambient temperature. The corona is generated in the packed-bed of barium titanate pellets or along the wire in the pulsed reactor. The necessity of heating the contaminated air streams to the temperature required for a catalyst or for thermal incineration to work is avoided.

The corona destruction processes were also evaluated as a means to control very low concentrations of contaminants in air streams. Experiments with contaminant streams using 10 ppmv single component VOCs in air demonstrated the ability to destroy the contaminant beyond the detection limit of our analytical equipment ( $< 10$  ppbv). Corona destruction may be an alternative control technology for low concentration streams where conventional control devices such as catalytic incineration and carbon adsorption have disadvantages, either economical or technical. Preliminary power estimates for the corona process indicate that the power requirement for VOC destruction is approximately  $3 \text{ W/ft}^3$  ( $106 \text{ J/sec/m}^3$ ). As shown in Figure 1, the annual operating costs of corona destruction fall below the costs of catalytic incineration and thermal incineration.<sup>2</sup> The annual operat-



**Figure 1.** Estimated costs of corona process and conventional VOC control technologies.

**Table II.** Potential source categories for the application of corona destruction.

#### Industrial

Painting and coating operations  
Semiconductor and electric component manufacture  
Food and pharmaceutical processing  
Site remediation

#### Commercial

Commercial paint operations (body shops)  
Furniture manufacturers  
Dry cleaning establishments  
Restaurants

ing costs for these technologies are based on a contaminated air stream at ambient conditions with a flowrate of 3000 acfm and an organic concentration of less than 100 ppmv. For these reasons, corona destruction may be well suited to destroy VOCs and air toxics in the outlet stream from operations such as the ones shown in Table II.

One of our current program objectives is to test the corona reactor on a larger scale. Experiments are currently underway to prove the feasibility of the packed-bed reactor on a 20 to 50 cfm ( $0.57$  to  $1.42 \text{ m}^3/\text{min}$ ) stream. At this stage, the reactor design can be optimized as to dimensions and power requirements. Other program objectives include demonstration of this technology on an industrial scale and commercialization of corona destruction as a VOC control alternative.

The purpose of this work is to determine if VOCs can be destroyed in these high intensity corona reactors. In setting up the original feasibility-of-concept experiments, an attempt was made to relate the work to the greatest extent possible to current problems threatening the environment. For these experiments both non-halogenated and halogenated compounds were evaluated.

### Theory of Corona Destruction

Although several theoretical concepts are still being evaluated to describe the destruction pathways for several families of hydrocarbons, the following discussion provides some of the most likely mechanisms for VOC destruction in a corona process.

Electrons undergo both elastic and inelastic collisions as they travel through energy fields. In an elastic collision, the electron retains the majority of its kinetic energy. Under the influence of the strong electric field, free electrons are accelerated. They undergo an elastic collision at the end of each free path length. The electrons continue to increase in energy until the energy becomes high enough to allow the electrons to undergo an inelastic collision. During an inelastic collision the electron transfers all or part of its kinetic energy to the particle with which it has collided. Inelastic collisions result in a change to the target particle or molecule such as ionization, dissociation or excitation.<sup>3</sup> In an inelastic collision, significant amounts of energy are transferred from an electron to the target species. Examples of these inelastic collisions are:

- Electron attachment by electronegative gases to form negative ions.
- Dissociation of molecular species into smaller fragments including formation of free radicals.
- Excitation of molecular and elemental species.
- Ionization to form positive ions and additional free electrons (a Townsend avalanche develops under favorable circumstances, generating many additional free electrons).



- Breaking down of molecular species into their elemental components.

The amount of energy required for the above events varies by type of event and molecular/elemental species. Energy requirements for different types of events are less than 5 eV for electron attachment and 5 to 25 eV to form positive ions by electron removal.<sup>4</sup> An electron volt (eV) is defined as the energy that an electron acquires (or loses) in passing through a 1 V change in potential. The electron volt is a particularly useful unit of measurement for this work because it allows an easy comparison of electrical energy input required for destruction of the target molecules. The probability of one of the above events occurring is expressed by the collision cross section which is mainly a function of concentration. The probability that the event will occur is dependent on the electron's having achieved the energy level needed for the event.

At atmospheric pressure and ambient temperatures, an electron's energy level can increase by a fraction of 1 eV in one mean free path length, if the mean free path length is parallel to an electric field of 20 to 30 kV/cm. At atmospheric pressure and ambient temperature a mean free path length is about  $1 \times 10^{-7}$  m.

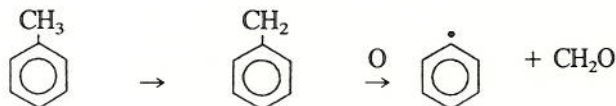
In addition to the above effects of inelastic electron collisions, there are also photoelectric effects (ionization, dissociation, excitation, etc.) which are either activated by or result in the emission of a photon. The events that can occur are very much interrelated.

In summary, the picture that is presented is one of high complexity when the possibility of electron collisions and the effects of photons are considered. The energy distribution in a swarm of energetic electrons ranges from very low to very high, with the majority centered around some median value. Therefore, electrons having a wide range of energies will undergo various types of inelastic collisions such as attachment, excitation and ionization. For a mixture of gases the picture becomes even more complex.

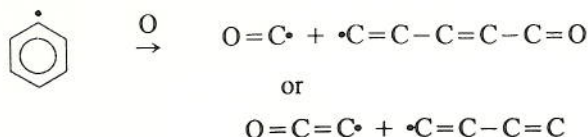
The power input to the bench-scale packed-bed reactor was measured, allowing the calculation of the energy introduced for a unit of time. For a typical toluene concentration of about 200 ppmv the energy introduced per toluene molecule was about 400 eV (9300 kcal/g-mole). The carbon byproducts were CO<sub>2</sub> and CO. The ratio of CO<sub>2</sub> to CO was about 2:1.

A number of reaction mechanisms are possible in the corona destruction of aromatic hydrocarbons. Three of the reactions are presented and discussed. The first reaction mechanism involves initial attack of the hydrocarbon molecule with an energized oxygen molecule. The other two reaction mechanisms, which involve breaking a C-C bond in the ring or removing a hydrogen from one of the carbons in the ring, have a lower probability.<sup>5</sup>

A mechanism likely to occur in the destruction of toluene is the oxidation of the methyl group of the molecule. Toluene has a resonance structure where a proton is lost or gained at the methyl group which should result in a more reactive site. The methyl group serves as an electron donor to the phenyl group. An excited oxygen molecule would attack the methyl group and the following reaction would proceed:



The CH<sub>2</sub>O radical rapidly reacts to form a CHO radical which goes to CO. The benzene radical reacts as follows:



The O=C or O=C=C radical reacts with oxygen to form CO<sub>2</sub>. The other radical oxidizes rapidly to CHO and then to CO.<sup>2</sup> Although the reaction to form CO rather than CO<sub>2</sub> is favored at low temperatures, high temperatures may be generated at the pellets, which would explain the favored formation of CO<sub>2</sub>. A CO<sub>2</sub> molecule is formed when the ring breaks, but the CO reaction is favored in the remainder of bond destruction reactions. This accounts for the approximately 1.7:1 ratio of CO<sub>2</sub> to CO observed experimentally.

The energies of bond formation/destruction are:<sup>3</sup>

C-C	3.6 eV
C=C	6.3 eV
C=C (in ring)	5.5 eV
C-H	4.3 eV
C-O	3.7 eV
C=O	7.7 eV
C=O (in CO <sub>2</sub> )	8.3 eV
C-Cl	3.5 eV

During the destruction of a toluene molecule, energy is required to break a C-H bond by an electron or by reaction with oxygen. The radical formed will react with an excited oxygen molecule, e.g., singlet oxygen (<sup>1</sup>Σ<sub>g</sub><sup>+</sup> and <sup>1</sup>Δ<sub>g</sub>) to form a C-O bond.<sup>6</sup> The energy released when the C-O bond forms is more than enough to break the adjacent C-C bonds. Therefore, the energy for the reaction once past the initiation energy must come from the oxidation of the toluene itself.<sup>7</sup> Because 20 percent of the gas is oxygen and since the oxygen molecule is one of the easiest to excite, the excitation of the oxygen molecule is consequently the most likely mechanism occurring in the process. The potential energies for the ground state and the first four electronically excited states of oxygen are shown in Table III.

The addition of energy greater than 7 eV causes dissociation of the oxygen molecule to one atom in the ground state and one in the 1st excitation state.<sup>8</sup> Many other excited states of oxygen are possible inside the corona destruction reactor.

The benzene molecule should react similarly to the toluene molecule, but slightly more energy would be required to initiate the benzene reaction. Instead of the excited oxygen attacking the methyl group of the toluene molecule, the point of attack in the benzene molecule would have to be the ring structure or a C-H bond. The bond energies would favor an attack of the C-H bond (4.3 eV for the C-H bond compared to 5.5 eV for the C=C bond). This is slightly

Table III. The potential energies for the excited state of oxygen.

State of Oxygen Molecule	Energy Required eV	Comments
Ground state	0	
1st excitation	0.98	Forbidden transition
2nd excitation	1.63	Allowed; half-life of about 10 seconds; collisionally relaxing to 1st excitation state
3rd excitation	4.25	Theoretically forbidden
4th excitation	6.00	Allowed; creates two oxygen atoms in the ground state via a non-radiative transition

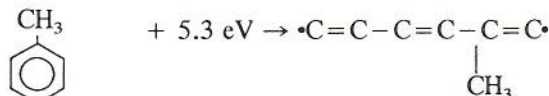


higher than the C-C bond energy which would be the point of attack in the toluene molecule (3.6 eV for the C-C bond). Therefore, since a higher energy is required to initiate the reaction of the benzene molecule, destruction of benzene should be lower than toluene under similar conditions. This was confirmed by our laboratory results.

The second possible reaction is the breaking of a C-C bond in the chain by a sufficiently energetic electron. This reaction is of the type:



in which [AB] is a radical. For the toluene molecule, the ring structure has a greater affinity for an electron than does the methyl group. The energy required to break a C-C bond in the toluene ring is 5.3 eV (123 kcal/g-mole). For toluene, cleavage of an arbitrary C-C bond would be:



The fragment on the right-hand side of the above reaction equation is a radical. The energy required for the ionization of toluene is 8.5 eV (195 kcal/g-mole). Consequently, the collision cross section for the C-C bond cleavage is considerably less than that for the ionization.

Once a C-C bond is broken and the free radical is formed, the free radical is able to react with oxygen. The heat of oxidation of toluene is 39 eV (901 kcal/g-mole) when going to CO<sub>2</sub> and 22 eV (497 kcal/mole) when going to CO.<sup>3</sup> Either pathway will supply sufficient energy to sustain the oxidation of all carbon molecules once the reaction starts.

For this second reaction to occur, electrons would have to achieve 5.3 eV to sever a C-C bond. Many electrons do achieve the higher energy levels, but not all.

A final reaction possibility is removing a hydrogen from the ring structure by electron collision. The energy required to break a C-H bond is about 4.3 eV. Once the C-H bond is broken the reaction will proceed by the same mechanism as the primary reaction suggested above. This mechanism would also account for the CO/CO<sub>2</sub> formation.

The intermediate steps in the oxidation require the attachment of an oxygen to a severed carbon bond or to a site where a hydrogen was removed. The energy released by attachment of the oxygen is sufficient to break an adjacent C-C bond, which provides the site for the next oxygen attachment. The most likely intermediate byproduct is the radical CHO which is favored at low temperatures. The CHO radical leads to the formation of CO; higher temperature reactions favor the formation of CO<sub>2</sub>. Note that once a C-C bond in the ring is severed, the radical that is formed has two active ends for attachment of an oxygen.

In all the suggested mechanisms, once the reaction of the individual molecule is initiated, the destruction of the molecule proceeds to completion since no other lower molecular weight species are found during analysis. If the molecule were not completely oxidized, other hydrocarbon byproducts would appear in the exhaust stream. The absence of other hydrocarbons has been confirmed by GC/MS. For operating conditions in which less than 100 percent destruction is deliberately achieved, the unreacted toluene molecules remain intact, which is evident in GC/MS.

## Results and Discussion

The packed-bed corona reactor makes use of a bed of ferroelectric pellets across which an AC electric field is impressed. A total of 10 hydrocarbons (benzene, cyclo-

hexane, ethanol, hexane, hexene, methane, methylene chloride, methyl ethyl ketone, styrene and toluene) were tested in the bench-scale packed-bed reactor. The bench-scale packed-bed reactor is depicted in Figure 2.

The pellets must be made of a material with a high dielectric constant. In this case the material is barium titanate with a dielectric constant ranging from 15 to 12,000.<sup>9</sup> The dielectric constant for barium titanate varies due to temperature, bead size and impurities. The barium titanate pellets are energized by an AC voltage applied through porous stainless steel plates. Corona appears at the contact points of the pellets when an AC electric field is generated as low as about 1 kV/cm. Sparking across the bed occurs for fields of 5 to 8 kV/cm, depending upon the size of the pellets. The reactor obviously uses more energy during sparking; however, the reactor performance is not improved proportionally to the increased amount of power applied during sparking. The most efficient operating point will be below sparking conditions.

The work required to remove a given electron from its atomic orbit and place it at rest at an infinite distance, is called the ionization potential.<sup>8</sup> Since all the compounds evaluated in the laboratory were not destroyed equally in the packed-bed corona reactor, the first method attempted to predict the destruction efficiency was based on ionization potential (see Table IV). Other parameters evaluated were heat of combustion, size of molecule, molecule bonding energy, Gibbs free energy and enthalpy. From the parameters evaluated to estimate the destruction of compounds in the packed-bed corona reactor, the ionization potential was the value that best correlated with destruction efficiency, i.e., the higher the ionization potential, the lower the destruction at constant operating conditions. As shown in Figure 3, factors other than ionization potential may explain the deviations from the predicted destruction efficiencies. In addition to the great ionization power of electrons, they also have the property of attaching to many molecular and atomic species to form negative gas ions. Electron attachment is greatest for atoms in the upper right-hand region of the periodic table. For example, methylene chloride has

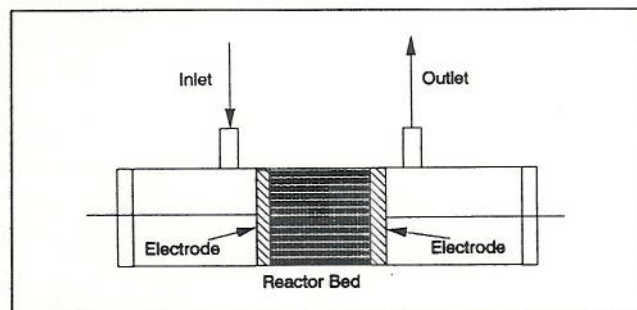


Figure 2. Bench-scale packed-bed corona destruction reactor.

Table IV. Ionization energies of selected molecules, eV.

Nitrogen	15.6	Formaldehyde	10.9
Hydrogen	15.4	Methanol	10.9
Carbon Monoxide	14.0	Hexane	10.2
Carbon Dioxide	13.8	Nitrogen Dioxide	9.8
Methane	13.0	Methyl Ethyl Ketone	9.5
Nitrous Oxide	12.9	Hexene	9.4
Ozone	12.8	Cyclohexane	9.4
Water	12.6	Benzene	9.3
Oxygen	12.1	Nitric Oxide	9.3
Methylene Chloride	11.3	Toluene	8.8
		Naphthalene	8.1



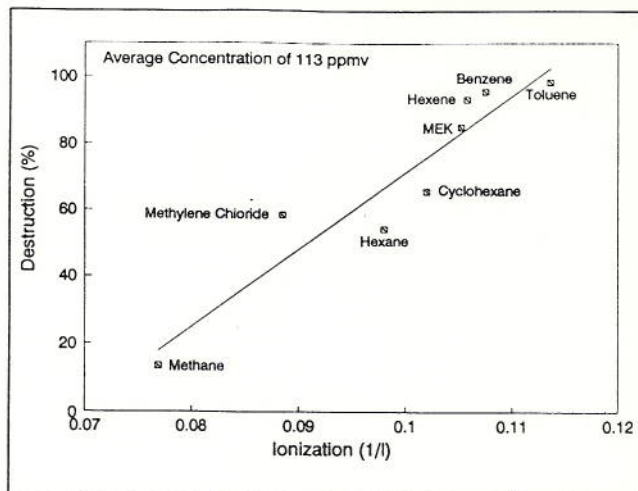


Figure 3. Destruction of VOCs as a function of ionization potential.

a higher ionization potential than hexane; however, the destruction efficiency is higher. This is probably due to the high electronegativity of the chlorine atom which will increase the chance of electron attachment to the chlorine atom.

A test matrix consisting of three sets of eight runs for a total of 24 runs was performed using toluene in the bench scale corona reactor. This test matrix was designed to support the calculation of material and energy balances, characterize wall effects, and determine the magnitude of systematic and random errors. Two reactor sizes (diameters of 2.23 and 3.18 cm), two face velocities (1.8 and 5.3 cm/sec), two residence times (0.48 and 1.43 seconds), and two toluene concentrations (50 and 250 ppmv) were used for the test matrix. The reactor length was 2.5 cm. The reactors were placed in an oven at 50 °C to maintain a constant temperature. Before each block of eight runs was initiated, the reactors were disassembled, the pellets were replaced by an equal weight of unused pellets, and the reactors were reassembled. Figures 4 and 5 show the outlet concentration of toluene, CO<sub>2</sub> and CO obtained during these experiments for toluene inlet concentrations of 50 and 250 ppmv, respectively. Each reactor performance was controlled by the gas flowrate (face velocity and residence time). The product of face velocity and residence time was maintained at about 2.5 cm for both reactors (R<sub>1</sub> and R<sub>2</sub>). The carbon balance for the test matrix varied from 102 to 105 percent.

In Figure 6, the effect of voltage on reactor performance for the large reactor (R<sub>2</sub>) and for a toluene concentration of 50 ppmv is shown. For voltages over 15 kV complete destruction of toluene was obtained.

The electric power for these experiments was measured using a digital oscilloscope connected to the corona reactor. The circuitry used to measure both voltage and amperage is depicted in Figure 7. Since the current signal is not of a sinusoidal form,  $ei \cos \theta$  was not used to estimate power consumption. Power was calculated by integrating the area under the power curve using Simpson's rule. The power curve is the result of multiplying each data point of the current curve with the corresponding data point of the voltage curve. The voltage and amperage for one cycle (16.7 ms) was analyzed using a customized spreadsheet program. Figure 8 is an example of the graphical output from the power curve analysis.

## Conclusions

The packed-bed corona destruction process shows significant promise as an alternative control method for re-

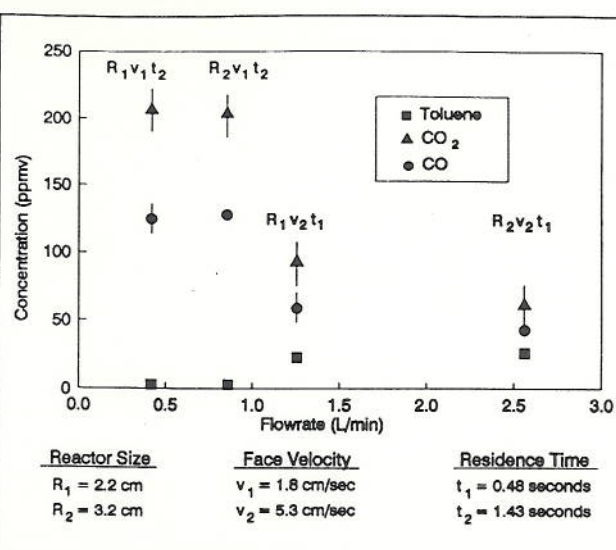


Figure 4. Outlet concentration of toluene, CO<sub>2</sub>, and CO at various flowrates (toluene inlet concentration of 50 ppmv).

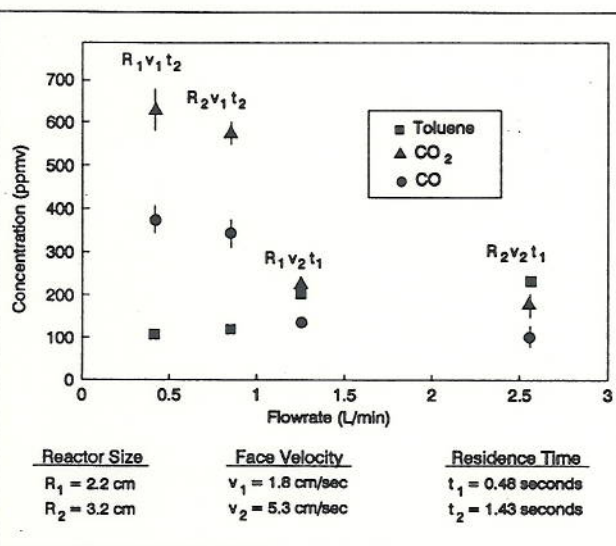


Figure 5. Outlet concentration of toluene, CO<sub>2</sub>, and CO at various flowrates (toluene inlet concentration of 250 ppmv).

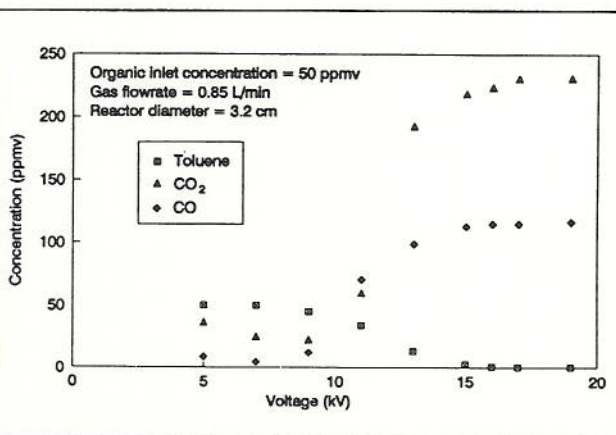


Figure 6. Toluene, CO<sub>2</sub>, and CO outlet concentration as a function of voltage.

ducing VOC and air toxics emissions from industrial and commercial operations. Preliminary data show that this technology is technically and economically feasible. When the corona destruction process is compared to conventional control technologies such as carbon adsorption, catalytic



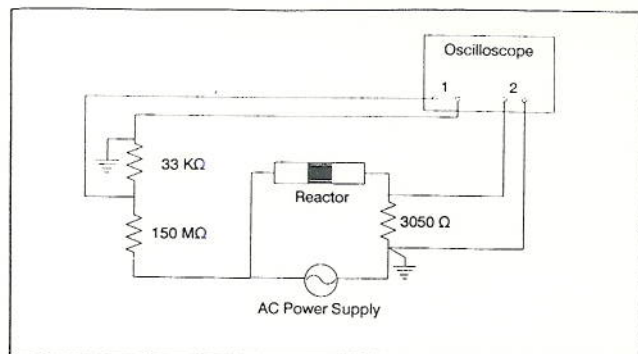


Figure 7. Voltage and current measurement system.

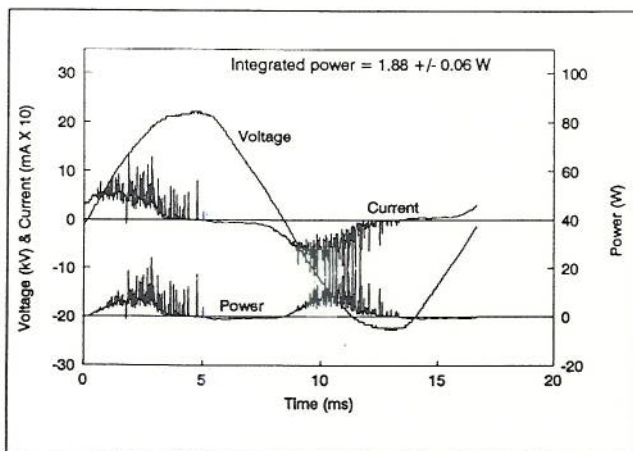


Figure 8. Power curve for the bench-scale packed-bed corona reactor.

incineration, and thermal incineration, the corona process demonstrates several significant advantages over the conventional control methods. These advantages are listed in Table I.

Conventional control technologies usually fail either technically or economically in effectively controlling VOC and air toxics at very low concentrations. In the corona destruction process, the lower the concentration the lower the power required to achieve excellent performance. Based on the durability of the bed packing material, ease of assembly, and simplicity of the hardware required, packed-bed corona destruction requires very little maintenance.

In catalytic and thermal incineration systems, relatively high temperatures are required to obtain the desired performance. This usually translates to a high cost of operation. In addition, when the inlet organic concentration is low, auxiliary fuel is needed to maintain proper incineration of the pollutants. However, the corona destruction process operates at ambient temperature, thus substantially reducing operating costs. Unlike catalytic systems, the corona process is not sensitive to poisoning by sulfur or halogen containing compounds.

Corona destruction also eliminates the problem associated with the treatment and disposal of the adsorbent used in carbon adsorption. Although the use of carbon adsorption in recovering VOCs may be cost-effective at high adsorbate concentrations, its performance dramatically declines when the adsorbate concentration is low. The application of carbon adsorption at low concentrations then becomes prohibitive.

Corona destruction is an effective alternative control method for a variety of VOCs and air toxics. Its destruction efficiency for VOCs such as benzene, cyclohexane, ethanol, hexane, hexene, methane, methylene chloride, methyl ethyl ketone, styrene and toluene may be predicted from the ionization potential and types of bonds in the molecules.

During the tests of destruction of VOCs and air toxics by the corona process, no products of incomplete reaction are formed. When the compounds were treated with the corona, molecules either were completely destroyed or passed through the reactor unaffected. More complex compounds and mixtures will have to be tested to determine if these promising results hold true in practical applications. The percentage of molecules destroyed can be predicted and controlled by appropriate reactor design and specification of operational parameters.

The performance of the corona destruction system can be enhanced by either increasing the power input or adjusting the residence time in the reactor to achieve a high degree of destruction for VOCs.

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