APPLICATION OF PULSED POWER AND POWER MODULATION TO THE NON-THERMAL PLASMA TREATMENT OF HAZARDOUS GASEOUS WASTES

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Introduction

Acid rain, global warming, ozone depletion, and smog are preeminent environmental problems facing the world today. Non-thermal plasma techniques offer an innovative approach to the cost-effective solution of these problems. Many potential applications of non-thermal plasmas to air pollution control have already been demonstrated.* The use of pulsed power and power modulation is essential to the successful implementation of non-thermal plasma techniques. This paper provides an overview of the most recent developments in non-thermal plasma systems that have been applied to gaseous waste treatment.

Nitrogen oxides (NO_x) and sulfur dioxide (SO_2), produced primarily by coal-burning power plants and automobile exhaust, are major contributors to acid rain. Carbon dioxide, released in large quantities when fossil fuels like coal, oil and natural gas are burned, is the biggest contributor to the accumulating greenhouse shield. Volatile organic compounds (VOCs), such as chlorofluorocarbons (CFCs) and halons, have been widely used in air-conditioning, refrigeration and aerosol products, and are present in fumes given off by gasoline, paints, and degreasing agents. They are used by the electronics industry as cleaning agents and by the chemical industry in the manufacture of foam products. VOCs are the main threat to the ozone layer, and contribute to chemicals that could lead to global warming. Soot particles and hydrocarbons emitted by cars and trucks are leading contributors to smog pollution. Well understood conventional technologies do exist for the treatment of some of these toxic gases, but they have practical limitations imposed by cost, energy requirements and byproduct disposal. Non-thermal plasma techniques offer the advantages of energy efficiency and the capability for the simultaneous removal of coexisting pollutants.

In devices such as plasma torches, the plasma is used only to create heat, which is applied indiscriminately to all the components of the waste. In the non-thermal plasma approach, the non-equilibrium properties of the plasma are fully exploited. These plasmas are characterized by high electron temperatures, while the gas remains at near ambient temperature and pressure. The energy is directed preferentially to the undesirable components, which are often present in very small concentrations. These techniques utilize the dissociation and ionization of the background gas to produce radicals which, in turn, decompose the toxic compounds. For many applications, particularly in the removal of gaseous pollutants, the non-thermal plasma approach would be most appropriate because of its high energy efficiency and its capability for the simultaneous removal of various pollutants. For others, including many mixed waste streams, the best approach might be to use a thermal plasma to incinerate the complete waste, including its container, and to use a non-thermal plasma reactor to clean the off-gases.

The key to success in the non-thermal plasma approach is to produce a discharge in which the majority of the electrical energy goes into the production of energetic electrons, rather than into gas heating. For example, in a typical application to flue gas cleanup, these electrons produce radicals, such as O and OH, through the dissociation or ionization of molecules such as H_2O or O_2. The object is to make acid rain in the flue gas instead of in the atmosphere. The radicals diffuse through the gas and preferentially oxidize the nitrogen oxides and sulfur oxides to form acids that can then be easily neutralized to form non-toxic, easily-collectible (and commercially salable) compounds.

Non-thermal plasmas can be created in essentially two different ways: by electron-beam irradiation, and by electrical discharges.

Electron-Beam Irradiation

Electron-beam irradiation has been proven to be an effective method for removing nitrogen oxides and sulfur dioxide in flue gases from industrial plants such as power stations and steel plants. In this method, the energy of the electron beam is used directly to dissociate and ionize the background gas. During the ionization by the beam, a shower of secondary electrons are produced, which further produce a cascade of ionization and dissociation. This

*On Sept. 21-25, 1992, a NATO Advanced Research Workshop on "Non-Thermal Plasma Techniques for Pollution Control" will be held in Cambridge University, England, to discuss laboratory studies and industrial implementations of non-thermal plasmas for the abatement of hazardous gaseous wastes.

cascading effect produces a large volume of plasma which can be utilized to initiate the conversions of NO_x and SO_2 to aerosols which can be collected by electrostatic precipitators or by bag filters. From the results obtained by basic studies and pilot-plant tests which have been done in Japan, Germany, Poland, and the USA, the process is considered to have an excellent potential for the simultaneous removal of NO_x/SO_2 without producing the reaction end waste products encountered in conventional absorption treatment. In Japan, the development of this process has been a result of the joint effort since 1970 by the Ebara Corporation (Kawamura et al. 1972), the Japan Atomic Energy Research Institute (Tokunaga et al. 1984), and Nippon Steel. In the USA, the evaluation of this process has been conducted by the Ebara Environmental Corporation (Frank et al. 1990) and Research-Cottrell (Helfritch et al. 1984), with support from the US Department of Energy. In Germany, pilot-plant studies have been performed at the Nuclear Research Center, the University of Karlsruhe, and Badenwerk (Willibald et al. 1990) (Platzer et al. 1990) (Jordan 1990). In Poland, a demonstration facility has been in operation at the Institute of Nuclear Chemistry and Technology in Warsaw. In Russia, several programs on electron-beam irradiation of flue gases are underway in Moscow at the Institute of Chemical Physics (Shvedchikov et al. 1988) and the Kurchatov Institute (Baranchicov et al. 1992).

Three advanced pilot plant tests are now being conducted for electron-beam treatment of flue gas in Japan (Maezawa et al. 1992). At the Ebara Corporation research facility in Fujisawa, the electron-beam process is being fine tuned for commercial use, and testing is being performed on incinerator gases and diesel truck exhaust gases. At the Chubu Electric Plant facility in Nagoya, the installation has its own boiler and can adjust the pollutant concentration of the flow gas, with studies focused on process optimization, equipment reliability, and byproduct handling. At the Tokyo Metropolitan tunnel facility, testing is being done to optimize a high-flow, low-NO_x-concentration exhaust gas treatment system in a vehicle tunnel under Tokyo Bay. In addition, the gases are analyzed for other hydrocarbons to determine the effects of the electron-beam process on unburned hydrocarbons. Electron-beam irradiation is now also being applied to the treatment of other types of hazardous emissions. In Germany, experiments are being conducted in Karlsruhe (Paur 1992) and by Asea Brown Boveri in Heidelberg (Esrom et al. 1992), to determine the removal efficiencies and energy consumption of electron-beam-induced cleaning of dilute emissions of volatile organic compounds.

The high capital cost of accelerators and x-ray hazard associated with electron-beam pollution control systems have motivated studies into alternate plasma-based technologies such as those utilizing electrical discharges. Electrical discharges can be produced in many different forms, depending on the geometry of the reactor and the electrical power supply. Many reactor designs use electrodes, such as small diameter wires, needles or sharp edged metals, that promote strong electric fields. The reactors are driven by direct current (DC), alternating current (AC), or pulsed power sources.

Pulsed Corona

One type of discharge reactor that has shown very promising results is the pulsed corona reactor. The industrial implementation of this reactor has the advantage of low retrofit cost since it can use the same wire-plate electrode arrangement as in electrostatic precipitators. Precipitators are commonly used for collecting particulate emissions in the utility, iron/steel, paper manufacturing, and cement and ore-processing industries. By driving the reactor with very short pulses of high voltage, short-lived discharge plasmas are created which consist of energetic electrons, which in turn produce the radicals responsible for the decomposition of the toxic molecules. Because of the short lifetime of the discharge, electrical power is not dissipated in the movement of ions, thus avoiding heating of the gas and providing good energy efficiency. Pulsed corona reactors have been shown, both in laboratory and industrial scale, to be very effective in the removal of many types of gaseous pollutants.

In Japan, experiments done in 1981 at the University of Tokyo investigated the possibility of enhancing the pollutant removal efficiency of the electron-beam method by applying an electric field to regenerate energetic electrons in the plasma (Masuda et al. 1981). It was found that pulsed electric fields could be very effective when a corona discharge was created, even when the electron beam was switched off. Subsequent tests at Masuda Research were performed in an incineration plant to apply this technique to the removal of other pollutants such as mercury vapour (Masuda et al. 1987). Recently, experiments conducted by Ishikawajima Harima Heavy Industries Co. and Toyohashi University of Technology, applied the pulsed corona method to the removal of ethylene for fresh fruit and vegetable storage (Kamase et al. 1991). A study was recently commissioned by the Japanese Ministry of International Trade and Industry to perform technical and economic assessments of the pulsed corona process for utility boilers. The committee concluded that the method deserves development as the next generation technology for the removal of SO_2 and NO_x, and estimated its costs to be 25% lower than that of the best existing technology.

Industrial-scale experiments on the use of pulsed corona for the simultaneous removal of NO_x and SO_2 from flue gas have been performed in Italy. The core of this project is a series of experiments carried out by the Italian National Electricity Board

(ENEL), in collaboration with the University of Padova, at the coal-burning power station in Marghera near Venice (ENEL 1988) (Dinelli et al. 1990). The ENEL tests have become the basis for the assessment of the electrical technology requirements of the pulsed corona process for applications to flue gas cleanup in actual power plants. The ENEL work has now evolved into a joint European Economic Community (EEC) project, with participation from Eindhoven University of Technology in the Netherlands and AEA Technology in the UK.

In the USA, laboratory experiments were conducted at Florida State University since 1983 that demonstrated the simultaneous removal of SO_2, NO_x and fly ash from effluent gases using the pulsed corona method (Clements et al. 1989). This work was supported in part by the US Department of Energy. The Research Triangle Institute has applied the pulsed corona method to the destruction of volatile organic compounds (Yamamoto et al. 1992). This program was started in 1985 under a US Environmental Protection Agency cooperative agreement, which included funding from the US Navy. Complete destruction was obtained for toluene, and high conversions were achieved for methylene chloride and trichlorotrifluoroethane (CFC-113). Research-Cottrell has applied the pulsed corona method to the decomposition of hydrogen sulfide (H_2S) (Helfritch 1991). H_2S is a toxic by-product of several industrial processes, including petroleum refining, dyeing, synthetic rubber, viscose rayon, and leather-treating operations.

Silent Discharge

In silent discharge reactors, AC high voltages are applied between electrodes, one or both of which are covered with a thin dielectric layer, such as glass. Silent discharges are also referred to as dielectric barrier discharges. The geometry is commonly either planar (parallel plates) or cylindrical (coaxial tubes). Configurations like those used in corona discharges are also used in which one of the electrodes (e.g. a wire) is highly stressed, and the outer electrode is a metal foil wrapped around a glass tube. Silent discharge processing is a very mature technology, first investigated by Siemens in the 1850's for the production of ozone. It is now routinely used to produce very large quantities of ozone for applications such as water purification, and the bleaching of textile and pulp.

Whereas in the pulsed corona method the transient behavior of the plasma is controlled by the applied voltage pulse, the plasma that takes place in a silent discharge self-extinguishes when charge build-up on the dielectric layer reduces the local electric field. For some applications, this feature presents an advantage for the silent discharge approach since simpler electrical power supplies can be used. In some cases, however, the efficiency of the silent discharge reactor is improved significantly by applying high-repetition-rate voltage pulses in a manner similar to that in pulsed corona reactors.

At Southern Illinois University, the plasma-assisted oxidation of SO_2 to SO_3 has been investigated using a silent discharge (Dhali et al. 1991). SO_3 is a more desirable byproduct of flue gas as opposed to SO_2 because it can be dissolved in water easily and can be reacted with lime to form gypsum. At the University of Illinois, the use of silent discharge processing for the removal of SO_2 from flue gas by conversion to sulfuric acid has been investigated (Chang et al. 1991). At the University of New Hampshire, the removal of NO has been investigated using a wire-cylinder silent discharge electrode configuration with a catalytic material, such as glass wool, packed in between the electrodes (McLarnon et al. 1991). The works at the Southern Illinois University, and the University of New Hampshire were funded by the US Department of Energy, while the work at the University of Illinois was funded in part by the US Environmental Protection Agency.

Silent discharge reactors have also been demonstrated to decompose many types of hydrocarbon, halocarbon, and organophosphorous compounds.

Mixed waste, containing both chemical hazards and radionuclides, are usually volume reduced through incineration and the residue packaged for long-term storage. The use of a silent discharge reactor as a second stage burner for removing hazardous organic materials from mixed waste is being investigated at Los Alamos National Laboratory (McCulla et al. 1991). Tests using a prepared gaseous feed containing trichloroethylene (TCE), carbon tetrachloride (CCl_4), n-octane and CFC-113 are encouraging.

The use of silent discharge processing for detoxifying chemical warfare gases has been investigated in several US laboratories. In 1985, work at the Naval Research Laboratory (NRL), funded by the US Office of Naval Research, demonstrated the decomposition of the simulant gas dimethyl-methyl-phosphate (DMMP) (Fraser et al. 1985). Collaborative work between Auburn University and the US Army Chemical Systems Laboratory demonstrated the decomposition of phosphonofluoride acid vapors (Clothiaux et al. 1984). Work at SRI International, with funding from the Wright-Patterson Air Force Base, studied the discharge-induced destruction of the simulants DMMP, DEMP, and O,S-DEMP (Slanger et al. 1992). The silent discharge-induced decomposition of methane, hydrogen cyanide, formaldehyde and benzene have also been reported in the experiments at NRL and Auburn (Fraser et al. 1985) (Fraser et al. 1986) (Neely et al. 1985) (Neely et al. 1988).

The US Army Chemical Research, Development and Engineering Center (CRDEC) has

recently conducted an evaluation of plasma technology for military air purification applications (Tevault 1992). Plasmas offer several advantages over the standard filtration approach, using activated impregnated carbon, in that all known military chemical and biological warfare agent can be decontaminated by passing through a plasma reactor. The CRDEC-sponsored technical evaluation of plasma air purification technology has identified several areas for further work required to mature plasma reactors for potential military application.

In Japan, collaborative work between Ibaraki University and Niles Parts Co. has resulted in the development of a near-practical plasma reactor that can simultaneously remove NO_x, SO_x, CO_x and soot from diesel engine exhaust (Higashi et al. 1992). This work began in 1979 following the failure of an effort to develop an exhaust gas recirculation (EGR) system to reduce NO_x emission from diesel engines. Although the primary goal was achieved, the recirculation of soot-laden gases led to decreased engine power and increased the wear on the valves, valve seats, piston surfaces and cylinder walls. In their cylindrical plasma reactors, the inner electrode was a stainless metal screw and the outer electrode was aluminum foil wrapped around a glass tube. The reactor was energized by AC high voltage. The major innovation in their work was the addition of drops of oil into the gas stream, which were transformed into a dielectric mist. This produced a more homogeneous discharge and led to the complete elimination of soot, as well as the removal of NO_x, SO_x and CO_x.

Surface Discharge

The use of surface discharge processing for the destruction of gaseous pollutants has been demonstrated in collaborative work between Masuda Research, University of Tokyo and Takuma Co (Masuda et al. 1985). The reactor consists of a planar or cylindrical alumina ceramic having a series of strip-like electrodes attached on one of its surface and a film-like counter electrode embedded inside the ceramic. A high-frequency AC high-voltage is applied to generate the surface discharge, starting from the side edges of the strip electrodes and uniformly covering the ceramic surface.

The surface discharge reactor has been applied to the removal of NO_x in the combustion gas from an oil-burning boiler and the exhaust from a diesel engine (Masuda et al. 1991). It has also been demonstrated that freons such as CFC-22 and CFC-113 can be destroyed without the production of poisonous by-products such as phosgene (Oda et al. 1991).

Ferroelectric Bed

The ferroelectric bed reactor employs a high-voltage AC power supply in conjunction with a tubular reactor packed with high-dielectric ceramic pellets. The pellets are held within the tube arrangement by two metal mesh electrodes. When external AC voltage is applied across the high dielectric layer, the pellets are polarized, and an intense electric field is formed around each pellet contact point. Many pulsed discharges take place around each contact point of the ferroelectric pellets, and the discharge energy can be controlled by changing the dielectric constant of the pellet, and by the voltage waveform.

At Toyohashi University of Technology in Japan, the ferroelectric bed reactor has been used to decompose methane diluted in various gas mixtures (Mizuno et al. 1988). In a joint work with McMaster University in Canada, experiments have been carried out on the reduction of carbon dioxide (Jogan et al. 1991). In the USA, experiments by Research Triangle Institute and the US Environmental Protection Agency have demonstrated that the ferroelectric bed can destroy a variety of hazardous organic compounds, including toluene, methylene chloride and CFC-113 (Yamamoto et al. 1992).

Microwave Discharge

The potential for using microwave discharges to destroy toxic molecules was demonstrated at the Lockheed Palo Alto Research Laboratory, in collaboration with Edgewood Arsenal, University of California at Berkeley, and the US Environmental Protection Agency (Bailin et al. 1975) (Bailin et al. 1978). Using microwave plasmas at sub-atmospheric pressures, they were able to destroy a wide range of hazardous molecules, including chlorinated hydrocarbons such as PCB's and organophosphorous compounds such as malathion. They also showed that metallic mercury can be recovered from the decomposition of phenylmercuric acetate (PMA), and performed an interesting series of experiments on the simulants DIMP and DMMP.

Experiments at the Kurchatov Institute in Russia (Bagautdinov et al.) and at Argonne National Laboratory in the USA (Harkness et al.) has demonstrated the use of microwave discharges for dissociating hydrogen sulfide. The plasma process has the advantage of being able to recover hydrogen, as well as sulfur, from the hydrogen sulfide.

Microwave systems for producing large volumes of non-thermal plasma in atmospheric-pressure gases have been proposed by investigators at AMPC Inc. (Guest et al. 1989) and the Naval Surface Weapons Center (Uhm et al. 1991).

Conclusions

Some of the critical issues restraining the commercial implementation of non-thermal plasma reactors are energy consumption and capital cost. These systems must operate with long life, require

minimal maintenance, and work well within limits related to the fraction of produced power available for the pollution control system. The pulsed power community has an important role in the development of high power switches, modulators, and accelerators that can lead to the commercial use of non-thermal plasmas for the solution of important environmental problems.

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