AIR POLLUTION CONTROL BY DC, PULSE AND MICROWAVE INDUCED NON-THERMAL PLASMAS

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(Accounted November 20, 2001)

Abstract. Several different plasma reactors were investigated for the air pollution control. An installation containing a DC negative corona discharge reactor, a pulse corona discharge reactor and a microwave induced plasma reactor is presented.

Key words: non-thermal plasma, SO\textsubscript{2}, NO\textsubscript{x}, removal efficiency, dc negative, pulse corona discharge, microwave, 2.45 GHz magnetron, reactors, pulse generator.

1. INTRODUCTION

Air pollution caused by gas emission of pollutants produced from a wide range of sources including coal, oil and gas burning plants, diesel engines, paper mills, steel and chemical production plants must be reduced drastically and urgently, as mandated by recent international agreements. Non-thermal plasma methods are very hot topics due to high pollutants removal efficiency, energy yields and good economy. Non-thermal plasma in which the mean energy of the electrons is substantially higher than that of the ions and the neutrals offer considerable advantage in reducing energy requirements to remove the pollutants. High energetic electrons induce molecule excitation, ionization and dissociation and at the same time, attachment of lower energy electrons to form negative ions at the discharge area. Secondary plasma reaction will be initiated by dissociated molecules, radicals and ions by radical and ion-molecule reactions in the downstream afterglow discharge region where the residence time is $10^{-3}$ to $10^{13}$ s. Also, aerosol particle surface reaction can occur and will depend on the composition of pollutant gas and additional reactant gases [1, 2]. Several different plasma reactors were investigated for the air pollution control. An installation containing a DC negative corona discharge reactor, a pulse corona discharge reactor and a microwave induced plasma reactor is presented.
2. METHODS, MATERIALS AND APPARATUS

2.1. METHODS

It is well-known that if an electron population with energies between 5 eV and 20 eV interact with a gas containing water vapor, oxygen, sulfur dioxide, nitrogen oxides and other gases produced by combustion of coal, oil, solid urban waste, etc., this will lead to ionization and excitation of the gas mixture components. If a sufficient amount of energy is transferred, excited molecules can decompose forming abundant quantities of active species, which are consumed in chemical reactions, part of them promoting the desired conversion of pollutants. Mainly, the oxygen atoms and radicals, such as OH, O₂H, react with NOₓ and SO₂ leading to their transformation into sulfuric acid and nitric acid. If ammonia is added to the gas mixture, the sulfuric acid and nitric acid are transformed into ammonium salts which can be removed by means of an electrostatic precipitator and used as fertilizers in agriculture. The electron population with energies between 5 eV and 20 eV may be obtained either by irradiating the gas mixture with high energy electrons produced by an external accelerator or by producing these electrons in the gas itself by subjecting all the gas to high electric fields [3-6]. DC corona discharge, pulse corona discharge and microwave induced non-thermal plasma, which is quite different from the thermal plasma, could be a source of high density of free electrons, which gives the possibility of obtaining a condition for chemical reactions under a mild temperature and atmospheric pressure. Thus, both external accelerated electron beam and non-thermal plasma can be regarded as means to generate free electrons and active species. The essential feature of combined DC, pulse discharges and microwaves could result in a decrease in the average power of the electron beam and a reduction in cost for the accelerators with the same removal efficiency. Some experimental studies show that the SO₂ removal efficiency can be increased by means of Argon in irradiated mixture as well as irradiation at lower temperature, below 70°C [7,8].

2.2. MATERIALS

The removal of NOₓ and SO₂ has been investigated in the following conditions: with 7% and without Ar added into gaseous mixture; NH₃ added as a stoichiometric quantity; SO₂ concentration up to 2000 ppm; NOₓ concentration up to 1000 ppm; CO₂ up to 10%; H₂O up to 25%; various MW power levels (up to 2kW); gaseous mixture flow rate of 1000lh⁻¹; mixture temperature of 65-70°C.

2.3. EXPERIMENTAL APPARATUS

The experiments are carried out using a flow system consisting mainly of the following units: a gas charging, mixing and controlling system; a gas analyzer; a DC negative corona discharge reactor supplied by a DC high voltage source; a pulsed
corona discharge reactor supplied by a pulse high voltage generator; a single mode microwave reactor. The analysis of the gaseous mixture composition is performed before and after treatment at the exit of the reaction reactors by means of a Portable Flue Gas Analyzer LANCOM 6500, supplied by LAND COMBUSTION, U.S.A. The schematic diagram of the experimental SO$_2$ and NO$_X$ removal installation is shown in Fig. 1.

The DC negative corona discharge reactor contains a ceramic tube injector (o.d. 3 cm and 10 cm long) with a hollow electrode (o.d. 0.5 cm and 5 cm long). A grounded ring type electrode (4 cm wide) is located on the outside surface of the ceramic tube. The DC corona discharge is generated by sharp hollow electrodes to the edge of the ceramic cylindrical tubes with a grounded ring electrode on the surface.

The pulse plasma discharge reactor is a coaxial one and consists of a centered single discharge wire of 1 mm in diameter and a stainless steel tube with 20 cm inner diameter and 250 cm length. The discharge wire is energized by short duration and fast rising positive high voltage pulses from a nanoseconds pulse generator. This generator was constructed to produce pulses up to 100 kV voltage, 1 $\mu$s duration and 200 ns rising time.

Two types of microwave reactors are used: with a multimode microwave cavity and with a monomode microwave cavity. The single mode microwave applicator is proper to be used to process poor absorbers of microwave energy. Briefly a single mode or monomode resonant applicator consists of a straight section cylindrical waveguide connected to a flange with a coupling iris on one side and a short circuit plunger on the other side. By varying the position of the
plunger the cavity can be made to resonate at the working frequency. The first microwave induced plasma reactor used in our experiments consists of an elongated cylindrical cavity particularly suited for the processing of temperature sensitive materials, including also low-loss substances. The microwave elongated cavity is adapted to operate in the TM$_{01n}$ mode and is provided with a waveguide propagating the electric field component of the microwave power substantially parallel to the axis of the chamber. The cavity ends are closed off with microwave chokes in order to minimize the energy radiation from the cavity. The microwaves are introduced into the elongated cylindrical cavity by a microwave injection system consisting of a microwave power controlled generator with 2.45 GHz magnetron of 850 W maximum output power, a rectangular waveguide launcher to fit to the WR430 waveguide, a dual directional coupler for forward and reflected power monitoring and a three stub tuner for impedance matching.

3. RESULTS AND DISCUSSION

The first experiments were carried out using a flow system consisting mainly of the following units: a gas charging, mixing and controlling system; a gas analyzer and an elongated cylindrical cavity. The air, SO$_2$, NO$_X$, Ar and CO$_2$ were individually charged and controlled using each flow-control valve and each flow-meter and than passed into the mixing chamber through each pipe to prepare the desired gases composition. The gaseous mixture prepared in the mixing chamber was charged into the reaction reactors at a flow rate up to 1 Nm$^3$h$^{-1}$. The temperature of the gaseous mixture was controlled by heaters set outside reactors. The photograph of the experimental gas charging, mixing and controlling system and of the microwave elongated cylindrical cavity is shown in Fig. 2.

The NO$_X$ or SO$_2$ removal efficiency, $\eta$(NO$_X$) or $\eta$(SO$_2$), is defined here as follows:

$$\eta$(SO$_2$) = 100\{C_i(SO_2)-C(SO_2)\}/C_i(SO_2)$;$$
$$\eta$(NO$_X$) = 100\{C_i(NO_X)-C(NO_X)\}/C_i(NO_X);$\n
where $C_i$(SO$_2$) or $C_i$(NO$_X$) are the pollutant concentrations before irradiation (initial concentrations) and $C$(SO$_2$) or $C$(NO$_X$) are the pollutant concentrations after applying microwave irradiation. The removal of NO$_X$ and SO$_2$ was investigated in the following conditions: with 7% and without Ar added into the gaseous mixture; NH$_3$ added as a stoichiometric quantity.

Fig. 3 shows the dependence of C(SO$_2$)C$^{-1}$(SO$_2$) ratio versus microwave irradiation time. This figure demonstrates that SO$_2$ removal is quicker and has a higher efficiency (from 68% to 77% ) using Argon.
The first experiments have shown that in the case of NO\textsubscript{X} removal the microwaves are not so effective in reducing the NO\textsubscript{X} concentration compared to SO\textsubscript{2} removal, which is up to 80% for 550 W. Some preliminary results demonstrated that NO\textsubscript{X} removal was facilitated when the MW treated gas was first passed through DC and pulse corona discharge reactors and by mixing various hydrocarbon additives in the gas mixture. Such experiments are now under investigation using the experimental installation shown in Fig. 1. The results will be further reported.
4. CONCLUSIONS

With an initial SO\textsubscript{2} concentration of 2000 ppm, a gas temperature of 65-70\textdegree{}C, a stoichiometric quantity of ammonia, a H\textsubscript{2}O concentration up to 25\%, a gas flow rate of 1000 l/h\textsuperscript{-1}, the results show that SO\textsubscript{2} is removed up to 80\% with microwave irradiation. The general conclusion from this study is that microwave treatment is a viable and promising method for flue gas cleaning, especially for SO\textsubscript{2} removal.

The Argon presence increases the SO\textsubscript{2} removal efficiency up to 20\%.

Using synergetic methods such as the combined methods with microwave irradiation and DC and pulse corona discharge, the removal efficiency of SO\textsubscript{2} and NO\textsubscript{X} grows up.

REFERENCES