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Comparative study of NO removal in surface-plasma and volume-plasma reactors based on pulsed corona discharges

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ABSTRACT

Nitric oxide (NO) conversion has been studied for two different types of atmospheric-pressure pulsed-corona discharges, one generates a surface-plasma and the other provides a volume-plasma. For both types of discharges the energy cost for NO removal increases with decreasing oxygen concentration and initial concentration of NO. However, the energy cost for volume plasmas for 50% NO removal, EC_{50}, from air was found to be 120 eV/molecule, whereas for the surface plasma, it was only 70 eV/molecule. A smaller difference in energy cost, but a higher efficiency for removal of NO was obtained in a pure nitrogen atmosphere, where NO formation is restricted due to the lack of oxygen. For the volume plasma, EC_{50} in this case was measured at 50 eV/molecule, and for the surface plasma it was 40 eV/molecule. Besides the higher NO removal efficiency of surface plasmas compared to volume plasmas, the energy efficiency of surface-plasmas was found to be almost independent of the amount of electrical energy deposited in the discharge, whereas the efficiency for volume plasmas decreases considerably with increasing energy. This indicates the possibility of operating surface plasma discharges at high energy densities and in more compact reactors than conventional volume discharges.

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1. Introduction

Atmospheric-pressure nonthermal plasmas with low gas temperatures, such as those generated by pulsed corona discharges and dielectric barrier discharges, are increasingly being used for air pollution control [1–3]. Such discharges are usually filamentary, comprising thin plasma channels (streamers) which propagate in the gas between the electrodes. Such plasmas are referred to as "volume-plasmas" in this report. The streamers can also propagate along solid–gas interfaces, as in surface-flashover, in creeping or sliding discharges [4,5]. They are referred to as "surface-plasmas" in this report. Interest in the application of surface-plasmas for environmental air pollution control is growing, particularly because of their energy efficiency which exceeds that of volume-plasmas [6–10]. This advantage in energy efficiency has been reported for the destruction of toxic volatile organic compounds (VOCs) [6,7], the oxidation of nitrous oxide (NO) to nitrogen dioxide (NO_2) [8,9], and the synthesis of ozone (O_3) [10]. The reason for the higher energy efficiency of surface-plasmas is assumed to be the more efficient mixing of reactive species, and the consequently higher reaction rates, due to the ionic-wind effect and surface mediated reactions.

Particularly environmentally harmful species are the nitrogen oxides (NO_x). They are present in diesel engine exhaust and in flue gases. About ninety percent of NO_x exist as NO. In order to remove NO from diesel exhaust or flue gases, it needs to be converted into NO_2 which is easier to remove. Removal of NO_2 can be achieved either by selective catalytic reduction [11–14], by adsorption [15,16], or by dissolution in water followed by reduction [17,18]. Oxygen based active species produced in the plasma, like ozone and atomic oxygen (O), are essential in converting NO into NO_2 [19,20].

Preliminary results under a limited set of experimental conditions show that the surface plasma is more efficient than the volume plasma for the conversion of NO into NO_2 [8,9]. It is desirable to test the surface plasma reactor under a broad range of experimental conditions. This manuscript reports the results of a more comprehensive study on the energy efficiency of surface-plasma and volume-plasma for NO conversion under a broad range of experimental conditions. The NO removal rate was measured as a function of the initial NO concentration and oxygen concentration, and on the specific input energy [21]. The study revealed new information that are of academic interest and important with respect of practical application of the technique.

2. Experimental

The schematic of the experimental setup is shown in Fig. 1. High voltage pulses of positive polarity (up to 30 kV and 500 Hz) were

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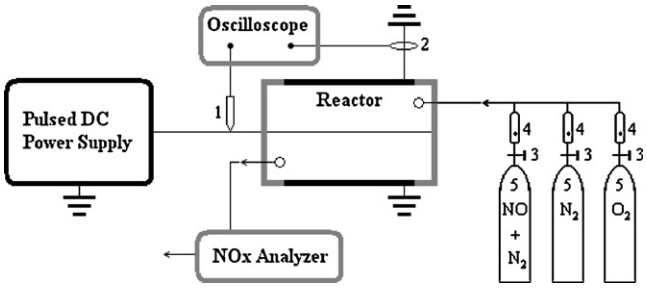

Fig. 1. Schematics of the experimental setup: 1 is the voltage probe, 2 current probe, 3 needle valves, 4 gas flow meters, and 5 pressurized gas bottles. The center line in the reactor depicts the wire anode and the bold lines on top and bottom, the two cathodes.

Table 1
Typical values of electrical parameters in this study.

Reactor	Parameter	Value
Volume plasma	Peak voltage	30 kV
	Voltage pulse width at half maximum	100 ns
	Voltage rise time (10–90%)	45 ns
	Energy per pulse	10 mJ
Surface plasma	Peak voltage	30 kV
	Voltage pulse width at half maximum	90 ns
	Voltage rise time (10–90%)	45 ns
	Energy per pulse	2 mJ

delivered by a "Compact Pulsed Power Modulator MPC3000S-OP1" (Suematsu Electronics Co., Ltd., Japan). The voltage and current diagnostics and energy calculations were the same as in our earlier study [6,9] using Tektronix TDS 3052 oscilloscope, Tektronix P6015A voltage probe and Pearson Electronics Current Monitor, Model 110A. The instant power was calculated from the product (VI) of the measured pulse voltage (V) and current (I). The energy per pulse (Ep) is the time integral ($\int VI\,dt$) of the power. The displacement current was measured by reducing the applied voltage to values below that required for the discharge breakdown or plasma formation. The typical values of the electrical parameters are listed in Table 1.

Two plasma reactors, i.e., a volume-plasma reactor (Fig. 2) and a surface-plasma reactor (Fig. 3), were employed in this study. The cylindrical electrode in the volume-plasma reactor was kept 2.5 cm away from end-fittings made of acrylic, which minimized the probability that a surface plasma would be generated along the end-fittings. The surface-plasma reactor comprises a wire to two parallel-plate electrodes stretched on the surface of a glass sheet

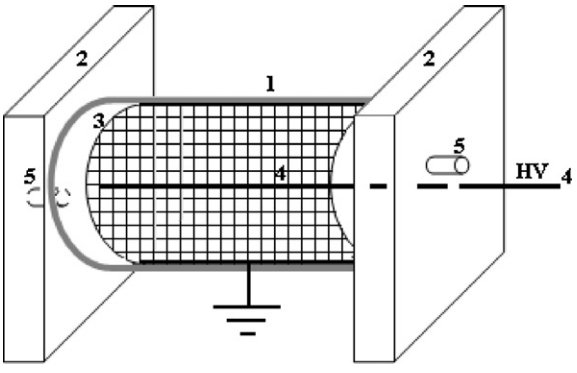

Fig. 2. Coaxial electrodes enclosed in an acrylic container were used for generating a volume-plasma. The components are: 1 is an acrylic cylinder of 4.5 cm ID, 5.1 cm OD, and 15 cm length; 2 are acrylic end-fittings; 3 is a cylindrical cathode of 4.5 cm OD, 10 cm length, made of stainless steel mesh; 4 is a stainless steel wire anode of 150 μm diameter stretched along the axis of the cylinder; and 5 are gas inlet/outlet.

with a second glass sheet parallel to the first one, with a spacer separating the two dielectric sheets.

Flow rates of nitrogen (N_2), oxygen (O_2) and NO from gas cylinders were controlled by needle valves and monitored with ball-float flow meters. Conditions of one atmospheric pressure, room temperature (25 °C), 1 liter per minute (L/min) flow rate, and 30 kV applied voltage were maintained in the experiments, except when mentioned otherwise. The concentration of oxygen and NO were monitored by an NO_x analyzer (ENERAC Model 500) equipped with oxygen and NO sensors. The resolution of the oxygen and NO sensors was 0.1%, and 1 ppm, respectively, and their accuracy, as specified by the manufacturer, was 0.2%, and 4% of the reading, respectively. Laboratory tests with nitrogen + oxygen mixture, NO + nitrogen, and NO + NO_2 + nitrogen mixtures show the accuracies for oxygen and NO measurements were well within the limits specified by the manufacturer.

In order to get stable inlet concentration of NO the process gases were allowed to flow for 1 h before the discharge was switched ON. The NO concentration at outlet was measured after 15 min of discharge ON. Three readings were averaged and each reading was an average of 3 min online monitoring. Error bars are not shown in figures because they were smaller than the size of symbols representing the experimental values.

Ozone (O_3) was estimated by two methods: (i) indirectly from the amount of NO consumed when the latter was mixed with the exhaust gases and (ii) directly by means of UV-spectroscopy using a UV-Ozone Analyzer model gFFOZ from In USA Incorporated.

The specific input energy (SIE) in units of Joule/liter (J/L) was calculated by using the formula:

$$\mathrm{SIE} = \frac{Epf}{Q} \tag{1}$$

where f is pulse frequency and Q is flow rate of process gas in liters per second (L/s).

The energy cost (EC) in units of electron-volts per NO molecule (eV/molecule) was calculated using the formula [22]:

$$\mathrm{EC} = \frac{250\mathrm{SIE}}{\mathrm{NO_{in}} - \mathrm{NO_{out}}} \tag{2}$$

where NO_{in} is the NO concentration at the inlet of the reactor and NO_{out} is the NO concentration at the outlet of the reactor, both in ppm.

The energy yield (EY) in units of the mass (in units of g) of NO removed per kWh input (g/kWh) is being calculated by using the formula:

$$\mathrm{EY} = \frac{1120}{\mathrm{EC}} \tag{3}$$

where the constant 1120 is calculated on the basis of the fact that $1\,\mathrm{eV} = 1.602 \times 10^{-19}\,\mathrm{J}$, $1\,\mathrm{kWh} = 3.6 \times 10^{6}\,\mathrm{J}$, $1\,\mathrm{mol} = 6.02 \times 10^{23}$ molecule, and 1 mol of NO = 30 g.

3. Results

A review of the scientific literature on volume-plasmas concludes that the NO removal rate is mainly determined by the specific input energy (SIE) [21]. The NO concentration and the corresponding energy cost values have therefore been expressed in terms of the SIE in the following figures. Fig. 4 shows that the NO concentration from air decreases with an increase in SIE. In this case the SIE was controlled by varying the pulse frequency and/or flow rate of the process gas. The energy cost was found to be almost independent of the flow rate, even for different pulse frequencies. In other words, the energy cost is only weakly, if at all, dependent on the pulse frequency under our experimental conditions.

The set of experiments with a flow rate of 1 L/min in a volume-plasma (circles in Fig. 4) was repeated (crosses in Fig. 4) to check

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