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Application of a non-thermal surface plasma discharge in wet condition for gas exhaust treatment: NOx removal

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ABSTRACT

This paper deals with the NOx removal with the help of a non-thermal surface plasma discharge in wet conditions. The gas treatment device consisting of a surface discharge and a wet-type reactor, was characterized through FTIR and electrical measurements. The ability of the proposed system for the cleaning of gas exhaust was studied. NOx as gaseous pollutant was decomposed effectively. To improve the chemical conversion, a coil was inserted in the electric circuit then a catalyst was placed in the plasma area. Results showed an improvement of NOx removal by an increase in radical species produced and synergistic effect, respectively.

1. Introduction

The control of air pollution represents a capital issue for the industry, and especially for the transport industry. Indeed, the increasing concern over the emission of air pollutants on the environment and human health has motivated the research to remove these harmful gases. To address this problem, several technologies to pollution control, environmentally acceptable and energy efficient, have emerged in recent years. Among these, the non-thermal plasma techniques offer an innovative approach to solve the problems of air pollution. The promising possibilities of non-thermal plasma processes (also called plasma reactors) for the elimination of pollutants and toxic molecules have already demonstrated, such as the removal of volatile organic compounds [1–3], sulphur oxides [4,5] and also nitrogen oxides [5–8].

In a non-thermal plasma, the mean energy of electrons is considerably higher than that of the other gas species. The highly energetic electrons produced by an electrical discharge, such as corona discharge or DBD, have a much higher probability to collide with neutral molecules of the surrounding gas (O_2 and N_2) than with the pollutant molecules because the content of pollutants is generally low. The collisions between neutral molecules and electrons produce active N and O species and OH radicals able to react with the polluted gases and to ensure their oxidations. In the case of NOx removal, the decomposition of NOx (the sum of nitric oxide (NO) and nitrogen oxide (NO_2)) by the non-thermal plasma process consists of the oxidation of NO by O to NO_2.

So, the NO is efficiently converted to NO_2, while the NO_2 can not be reduced effectively by N_2. Consequently, the non-thermal plasma technology presents limitations. To overcome this limitations, several researchers have used a wet-type plasma reactor [9–14]. The underlying process is the dissolution of NO_2 into water and its conversion to nitrate (NO_3^-) and nitrite (NO_2^-) ions.

In addition, most investigations on pollution control using a non-thermal plasma reactor are based on volume discharge. Alternatively to the volume discharge, a way to treat the exhaust gases consists of using a surface discharge. Still, this type of discharge is under employed in the field of the pollution control [15,16], while it has known an important development over the past fifteen years in the aerodynamic field [17,18].

This study focuses on the removal of NOx using a wet-type reactor combined to a non-thermal surface plasma discharge. The first part of this paper discusses the ability of the present system (*i.e.* reactor using a surface discharge) to clean the gas exhaust. In a second part, we show the experimental results about the effect of a coil inserted in the electric circuit on the treatment process. In a third part, we highlight a synergestic effect between the plasma and a catalyst (here γ-Al_2O_3). And in a last part, a comparison

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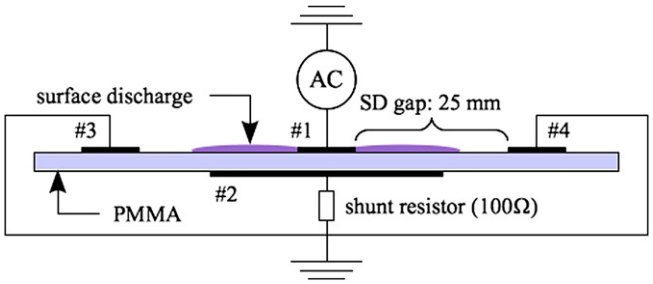

Fig. 1. Schematic illustration of the electrodes design with an electric circuit used in the experiment.

between the present results and plasma reactors from the literature is realized.

The three contributions listed above are analyzed by measuring the power injected into the gas, and comparing the variation in concentrations of NO and NOx through FTIR measurements. In addition, all experiments are conducted at low and constant flow rate (1 L/min), with an initial content of 100 ppm NO.

2. Experimental setup

2.1. Non-thermal surface plasma reactor

A schematic illustration of the non-thermal surface plasma discharge setup is shown in Fig. 1. The plasma device consists of two electrodes (electrodes #1 and #2) flush mounted on each side of a dielectric barrier, plus two counter-electrodes (electrodes #3 and #4) placed on the top side of the insulating wall. These counter-electrodes are separated relatively to the electrode #1 by an air gap of 25 mm, named the SD gap. Each electrode is made of 50-μm thick aluminium strip tape whose ends are oval in order to reduce edge effects. Upper electrodes are 60 mm-long (in spanwise direction) and 10 mm in width. The lower one is 30 mm wide for 60 mm-long. The dielectric barrier used is a polymethyl methacrylate (PMMA) plate of 140 mm × 100 mm and 3 mm-thick, with a permittivity ε_r equal to about 3.3 at 1 kHz and a dielectric strength of 15 kV/mm.

The surface discharge is powered by an AC HV power supply connected to the electrode #1, as shown in Fig. 1. The AC HV power supply is obtained with the help of a transformer supplied by a power amplifier (NF Corporation, model 4510, 1.25 kV A). The transformer may supply a maximum peak voltage of 20 kV at driving frequencies up to 5 kHz. The three other electrodes are grounded. In this case, the plasma device operates in the Dielectric Barrier Discharge (DBD) mode [19,20].

Fig. 2 displays a schematic side-view of the non-thermal surface plasma reactor used in this study. It consists of two parts. The first part consists of a reactor with two openings that allow the inlet and outlet gas. This reactor made of polypropylene (PP) has a rectangular cross-section 100 mm × 30 mm for 150 mm-long. The last part of the plasma reactor corresponds to the surface discharge (described above) located on the upper side of the reactor. The whole surface discharge and reactor forms the non-thermal surface plasma reactors.

The wet condition is obtained by adding a sodium sulfite (Na_2SO_3) solution, concentration of 1 mol/L, inside the reactor (see Fig. 2). This solution is used as NOx gas absorbent. Indeed as reported in the literature [9–14], a plasma reactor operating in wet conditions can improve the efficiency of NOx removal by dissolving NO_2 into the liquid as NO_2^- and NO_3^- ions. However, the continuous absorption of nitrogen oxides induces saturation and acidification of the liquid, resulting an inhibition of further absorption. The adding of sodium sulfite allows to reduce the nitrite and nitrate ions to N_2, thus the gas absorption is facilitated [21]. Moreover the aqueous solution of Na_2SO_3 was replaced after set of experiments.

Here, 150 mL of Na2SO3 solution was used. Thus, due to the size of the reactor, the height of 150 mL solution is 10 mm. In addition, taking into account the height of the reactor (30 mm) and the thickness of the PMMA plate (3 mm), the distance between surface plasma discharge and water is 17 mm.

2.2. Electrical measurements

One of key parameters to evaluate a pollution control system is the energy consumption. This one is determined from the simultaneous measurements of the discharge current and the applied voltage. The discharge current, i, is deduced from the voltage across a non-inductive resistor (100 Ω) connected in series between the electrode #2 and the earth, as shown in Fig. 1. The AC voltage applied to the electrode #1 is measured by using an HV probe (Tektronix, model P6015A, 3 pF, 100 MΩ). Each electrical waveform is recorded using a fast digital oscilloscope (Tektronix, model DPO 2024, 200 MHz, 1 GS/s).

A typical example of the voltage and current waveforms is given in Fig. 3, for a 3 mm-thick plasma device made of PMMA, a frequency of 1 kHz and a peak voltage of 12 kV. It can be observed that a series of short and intense current pulses occurs during the positive voltage half cycle, whereas only a few peaks of similar intensity are observed during the negative voltage half cycle.

From both voltage and currents curves, the total electrical power consumption can be calculated as follows:

$$P = \frac{1}{nT}\int_{t=0}^{nT} v(t)i(t)\mathrm{d}t$$

where $v(t)$ and $i(t)$ are the measured voltage and current versus time, respectively, T is the waveform period, and n is the number of periods. In practise, the time-averaged value computed with only

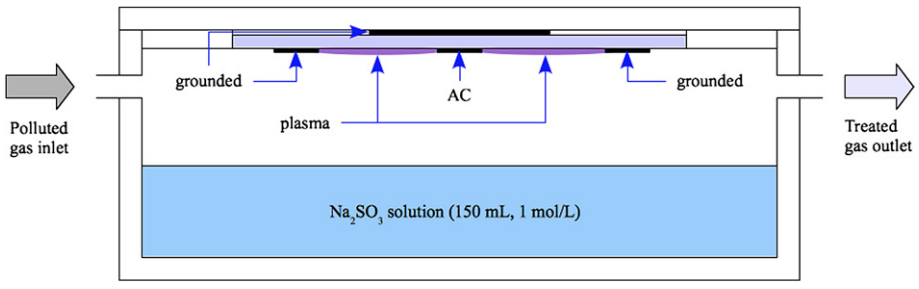

Fig. 2. Schematic side-view of the wet-type plasma reactor used.