NOx removal using a wet type plasma reactor based on a three-electrode device

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Abstract. In this paper, a wet type plasma reactor based on a three electrode device is investigated experimentally in order to remove NO and NOx at low flow rate. First, a comparison of cleaning performances of gas exhaust has been performed when the surface discharge operates in DBD or SD modes. From these previous results, the second part of study has consisted to improve the electrochemical conversion of the wet type plasma reactor by adding a coil between the AC HV power supply and the surface discharge. The parametric study has been performed with 100 ppm of NO content in gas flow at room temperature and atmospheric pressure for a flow rate of 1 L/min. For each electrical parameter tested, an electric characterization and measurement of NOx content via FT-IR has been conducted. The results highlight a better cleaning of gas exhaust when the surface discharge operates in DBD mode. Moreover, the presence of solution promotes the arc transition when the operating mode is SD, resulting a reliability reduction of plasma device. In addition, the measurements show that the insertion of coil in the electrical circuit improves the NOx removal at a given power consumption for the DBD operating mode.

1. Introduction

Pollution control represents a capital issue for the transport industry. Indeed, the increasing concern over emission of air pollutants on environmental and human health has motivated the research to remove these harmful gases. Therefore, several pollution control technologies, environmentally acceptable and energy-efficient, have emerged in recent years. Among these, the plasma devices (also called plasma reactors) appear as being suitable to reduce pollution and especially to remove NOx [1-5]. NOx removal of gas exhaust via non thermal plasma reactor is divided into dry and wet way. Several studies have shown that the wet type reactor allows to obtain significant results for gas exhaust cleaning compared to the dry reactor. The underlying process is the NOx dissolution into water and its conversion to nitrate (NO$_3^-$) and nitrite (NO$_2^-$) ions [1].

The objective of this work is to define an optimal configuration in terms of electrochemical conversion efficiency and reliability, of a wet type plasma reactor.

In this paper, we present a study on NOx removal performed via a wet type plasma reactor based on a three-electrode device. Depending on the applied electrics parameters [6, 7], the surface discharge shape (i.e. three-electrode design) enables to select the operating mode of the discharge. Each discharge regime is stable at room temperature and atmospheric pressure with a standard relative humidity (i.e. 40%). In a first part, the performances of NOx removal obtained by using the DBD and SD operating modes are shown. In a second part, effect of a coil inserted between the AC HV power supply and the plasma device (in DBD mode) on polluted gas is characterized.

2. Experimental setup

Figure 1 shows a schematic illustration of the discharge setup. The plasma device consists of two electrodes flush mounted on each side of a dielectric plate, plus two counter-electrodes 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 SD gap. Each electrode is made of aluminium strip whose ends are oval
in order to reduce edge effects. Upper electrodes are 60 mm-long (in spanwise) for 10 mm in width.
The lower one is 30 mm wide for 60 mm-long. The dielectric used is a PMMA flat plate (140 × 100
mm$^2$) of 3 mm-thick.

The bottom electrode is grounded when the electrode #1 is connected to an adjustable AC HV
power supply. The AC power supply is obtained with the help of a transformer supplied by a power
amplifier (NF Corporation, model 4510). The transformer can supply a maximum peak voltage of 20
kV at driving frequencies up to 5 kHz. Depending on discharge operating mode, the two other
electrodes are either grounded, corresponding to DBD mode, or connected to a negative DC power
supply (Matsusada Precision, model HAR-50R6), i.e. SD mode.

The AC voltage applied to electrode #1 is measured by using an HV probe (Tektronix, model
P6015A). Electric currents flowing through the system are deduced from voltages across shunt
resistors of 100 $\Omega$ connected to a fast digital oscilloscope (Tektronix, model DPO 2024). From voltage
and current curves, the total electrical power consumption can be calculated.

Figure 1. Scheme of plasma device used: DBD (left) and SD (right) modes.

Figure 2 displays a schematic side-view of the wet type plasma reactor used in this study. It is
composed of two parts. The first part consists of a wet reactor with two openings allowing the inlet and
exhaust of gas flow. This reactor made of polypropylene plate has a rectangular cross-section (100 × 30
mm$^2$) for 150 mm-long. The liquid inside the reactor is a sodium sulfite solution (150 mL, 1 mol/L)
which is used as NOx gas absorbent. Moreover, it allows to re-established the pH level by reduction
NO$_3^-$ ions to N$_2$ or NH$_3$. The second part of the plasma reactor corresponds to the surface discharge
placed on the top plate of the reactor. The whole surface discharge and wet reactor forms the wet type
plasma reactor.

The experiments are carried out at room temperature and atmospheric pressure. Total gas flow rate
is regulated to 1 L/min by the mass flow controller. The inlet gas composition is NO (100 ppm) and
dry air. NO and NOx concentrations (with [NOx] = [NO] + [NO$_2$]) are measured using FT-IR
spectroscope gas analyzer (BEST SOKKI, model SESAM 3-N).

In the cleaning experiment, the discharge is ignited twice 3 min, and this 3 min after the start of
recording of gas exhaust concentration. Each ignition is spaced 3 min, bringing the total duration of
experiment at 15 min. Moreover, the determination of average concentration is not achieved when gas
concentration reaches a steady state.

Figure 2. Schematic side-view of the wet type plasma reactor used
3. Results

3.1. NOx removal as function of discharge operating mode

In this section, we focus on the ability of the plasma device to remove NOx depending on the operating mode of the discharge. The initial NO content is fixed at 100 ppm and the polluted air flow rate is equal to 1 L/min.

First, we study the cleaning of gas exhaust when the surface discharge operates in DBD mode. In this case, the waveform applied is a sine one, at frequency set to 1 kHz. The peak voltage is between 6 and 13 kV. Figure 3 shows the evolution of NO and NOx concentrations versus the power consumed. For both gases, we can see that the initial content of pollutants is decreased with the injected power. The NO and NOx removal reaches 80% and 40%, respectively, for a power consumed about 2.25 W, corresponding in our case to an energy density of 135 J/L.

However, the computation of the average concentration is not realized during the steady state, resulting in a bias on the final value of the pollutant content. For example, the minimum concentration of NO in the steady state is about 7 ppm. In the case, the NO removal efficiency reached 93%.

The removal of NO proceeds mainly via oxygen species (O and O$_3$) in air, leading to the formation of NO$_2$. In the presence of the solution (wet condition), NO and NOx concentrations decrease due to the absorption of NO$_2$ into the liquid and/or reaction of NO$_2$ with OH radicals to forms NO$_3$ ions.

Figure 3. NO and NOx concentrations versus power consumed by plasma device in DBD operating mode.

Figure 4 presents the evolution of NO and NOx concentrations versus the power consumed in SD mode. Here, the experimental setup is the same as above, except the AC voltage applied equal to 7 and 8 kV peak. Moreover, the negative DC component is ranging between 0 to -11 kV. From this figure, several results appear. First, one can see that at given electrical power consumption the plasma device seems less effective for cleaning exhaust gas compared to the previous case. For example, at 1 W the minimum concentration of NO is about 75 ppm with the SD mode while it reaches 62.5 ppm with the DBD actuation. Then, we observe that the NO and NOx concentrations as a function of power consumed have an abnormal evolution. Indeed, we see that for an electric power range between 0.5 and 0.85~0.9 W, the injected power into the discharge does not improve the treatment of pollutants. On the contrary, outside this range a decrease of NO and NOx content is obtained.

However, it has not been possible to verify the phenomenon with others electrical parameter because the presence of the solution inside the reactor promotes the transition to arc of the discharge.

3.2. Effect of a coil on NOx removal

In this section, we study the influence of a coil on gas exhaust treatment. To achieve this, a coil is placed between the AC HV power supply and the plasma device. The NO concentration is fixed at 100 ppm and the polluted air flow rate is equal to 1 L/min.
As reported in previous section, the SD operating mode is unstable (transition to the arc) due to the presence of Na$_2$SO$_3$ solution. Therefore, we study the cleaning of gas exhaust when the surface discharge only operates in DBD mode. In this experiment, the waveform applied is a sine one with a peak voltage ranging from 6 to 12 kV, at frequency set to 1 kHz. Figure 5 highlights the NO and NOx concentrations as a function of power consumed without and with coil. It appears clearly that the coil integrated in the electrical circuit allows to improve the cleaning of gas exhaust, except the first measurement point. For example, at 1 W the NO concentration reaches 35 ppm in presence of the coil, whereas the NO content is only 65 ppm with the standard case (i.e. without coil). If we compare the NOx content in presence or absence of coil, we remark that the concentration difference is less significant, i.e. 65 ppm with coil against 80 ppm. It means the inductance allows a higher radicals production, resulting an effectiveness improvement of the NO treatment process. Similarly, it promotes the absorption of NO$_2$ in the solution of sodium sulphite, which leads to a decrease in NOx content.

The lower efficiency of gas exhaust cleaning for the first measurement point can be explained by the fact that the coil is resistive, about 4 kOhm/H, so the voltage applied across the plasma device is lower compared to the case without coil.

Figure 5. NO and NOx concentrations versus power consumed by plasma device without and with coil ($L=1$ H).

4. Conclusion

In this paper, we have shown the capability of a surface discharge to clean gas exhaust with 100 ppm of initial NO content for a flow rate of 1 L/min. Depending on our experimental conditions, it appears that a better cleaning of gas exhaust is obtained when the surface discharge operates in DBD mode. In addition, the evolution of pollutant content presents an abnormal evolution during the SD mode, requiring further investigation. However, the presence of the solution promotes the arc transition of this operating mode, resulting a reliability reduction of plasma device and prevents the use of others electrical parameter.

And finally, adding a coil in the electrical circuit improves the efficiency of gas exhaust treatment by increasing radicals production and promoting NO$_2$ absorption into Na$_2$SO$_3$ solution.

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