# Design Factors for $NO_X$ Reduction in Nitrogen Plasma

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***Abstract*—The aim of this paper is to analyze the influence of plasma discharge reactor unit geometries on the efficiency of dry $NO_x$ reduction process in nitrogen plasma environments. The experimental setup consists of the versatile plasma discharge reactor unit powered by 10-kHz pulse HV supply and supplied by mass flow controllers with a simulated $(NO + N_2)$ gas under different concentrations and flowing rates. The measurements are obtained by means of a gas analyzer for $NO/NO_x$ concentration and a digital oscilloscope supplied with HV and current probes for discharge power computation. The number of surface discharge electrodes, the distance between them, their active area, the gas flow rate, and concentration are studied while aiming at 30 $g(NO_2)/kWh$ energy efficiency and higher reduction efficiencies. These targets are the keys to industrial application of plasma discharge reactor, as a way of reducing $NO_x$ from the regeneration phase in the next generation diesel exhaust aftertreatment system, and the results proved successful.**

***Index Terms*—Diesel exhaust aftertreatment system, nitrogen plasma, $NO_x$, reactor geometry surface discharge.**

## I. Introduction

ONE of the major problems associated with internal combustion engines is the noxious exhaust emissions. Particulate matter (primarily carbon) and oxides of nitrogen such as nitric oxide (NO) and nitrogen dioxide $(NO_2)$ often collectively referred to as $(NO_x)$ are two of the most harmful materials, particularly in the case of diesel engines. Severe emissions control regulations are forcing vehicle manufacturers to find more efficient ways of removing these materials from internal combustion engine exhaust. One way to reduce emissions is by modifying the combustion process [1]: altering injection timing, engine design, common rail systems, and exhaust gas recirculation. In practice, it is found that combustion techniques which improve the situation in relation to one of the harmful components tend to worsen the situation in relation to others. There are numerous aftertreatment techniques being developed to remove $NO_x$ emissions from exhaust gases but, in general, practical $NO_x$ reduction systems are reliant on passing the exhaust gases across a catalyst using one of the two types of catalytic reduction methods: nonselective [2], [3] and selective catalytic reductions (SCR) [4]. The use of exotic catalysts and a constant supply of ammonia are amid several limitations preventing either of these to get ground in practical applications. Plasma chemical processing is a promising technology for harmful air pollutants' decomposition as thousands of ppm can be decomposed with reduced organic byproducts with only seconds of residence time. The suppression of inorganic exhaust products such as nitrogen oxides requires improved reactor geometry and control of input power and residence time.

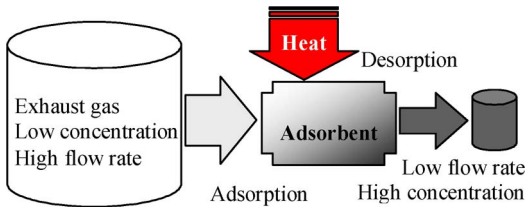


Fig. 1. Concept of concentrating exhaust gas using adsorption and thermal desorption.

A diesel emissions control system [5]–[7] currently in prototype stage has been using $NO_x$ adsorption, thermal desorption, and subsequent reduction by nonthermal plasma (Fig. 1). The $NO_x$ is first removed by adsorption from the high flow rate low-concentration exhaust gas, then the adsorbent is regenerated by thermal desorption of a low flow rate high-concentration gas using waste heat of the exhaust gas. In the regeneration process, hot exhaust gas passes through the heat exchanger tubes surrounding the adsorbent pellets. This system does not use a catalyst or special reducing agent like $NH_3$ but uses an oxygen poor gas. The main benefit of this technology comes from the fact that no fuel distribution infrastructure changes are needed. The desorbed NO is subsequently reduced to $N_2$ by nitrogen nonthermal plasma (NTP) $NO_x + N^* \rightarrow N_2 + xO$ ($N^*$ is the excited nitrogen atom) [8]–[10] generated by surface discharge in a reactor. This is fundamentally different from most approaches to $NO_x$ removal from diesel exhaust where NTP is used as a way to induce the gas phase oxidation of NO to $NO_2$ and assist in catalytic reduction (SCR or NSCR).

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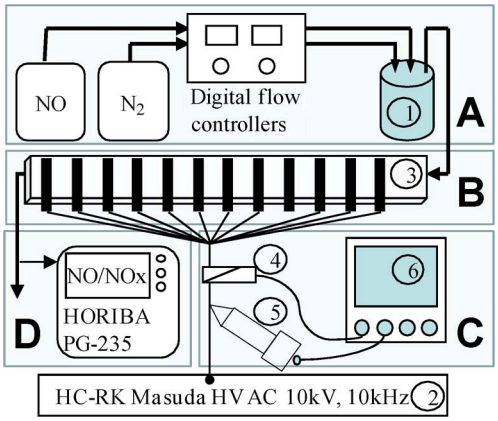


Fig. 2. Schematic representation of the experimental setup: (1) Gas mixing chamber; (2) HC-RV Masuda Research HV supply; (3) Plasma reactor; (4) Tektronix P6021 current probe; (5) IWATSU HVP60 HV probe; (6) Yokogawa DL1740 digital oscilloscope.

The NTP reactor consists of 12 surface discharge elements made of ceramic tubes. Each element generates NTP on the surfaces. Elements can be independently disconnected to study the interaction between their numbers, distribution, discharge power, reduction efficiency, and energy efficiency. The electrodes are powered by a 10-kHz HV supply with voltage & current controls. Gas flow rate is set by the mean of digital flow controllers and NO concentration before and after the reduction process is monitored using a NO/$NO_x$ gas analyzer. Based on industrial requirements, energy efficiency target was set at 30 g$(NO_2)$/kWh with an aim at high reduction efficiency. For industrial application, it is essential to fully acknowledge the effect of each design factor on both reduction and power efficiencies of the NTP reactor. The $NO_x$ reduction module discussed in this paper is a key component on the broader project "Development of Plasma Combined Exhaust Gas Aftertreatment System for Super Clean Diesel Hybrid Engine."

Simulated gas parameters were determined from real diesel exhaust data and the adsorption/thermal desorption mechanism in a cyclic adsorbent regeneration process [11]. A custom experimental setup was prepared for the independent development of this module. The equivalent of 20 L/min of poor oxygen gas containing about 2600 ppm NO must be treated according to thermal desorption output. While NO desorption benefits from higher gas flow, the impact on the reduction process requires assessment. Using simulated gas [12] with controlled NO concentration and constant flow rate, this paper will analyze the influence of NTP reactor geometries on the efficiency of $NO_x$ reduction process.

## II. Experimental Setup

The four main components of the experimental setup shown in Fig. 2 are gas simulation module, plasma reactor unit, discharge power measurement unit, and gas analyzer. The experimental setup allowed efficient independent research to be conducted on the NTP reactor in the development phase of the plasma combined exhaust gas aftertreatment system.

In order to prepare the simulated gas, 2% NO balanced with $N_2$ in a cylinder was mixed with 99.99% $N_2$ from a second cylinder. The desired concentration and flow rate were obtained using a gas mixing chamber (80 $cm^3$) and digital mass flow controllers (SAM, Hitachi Metals: 1, 2, 5, and 20 L/min) on each line.

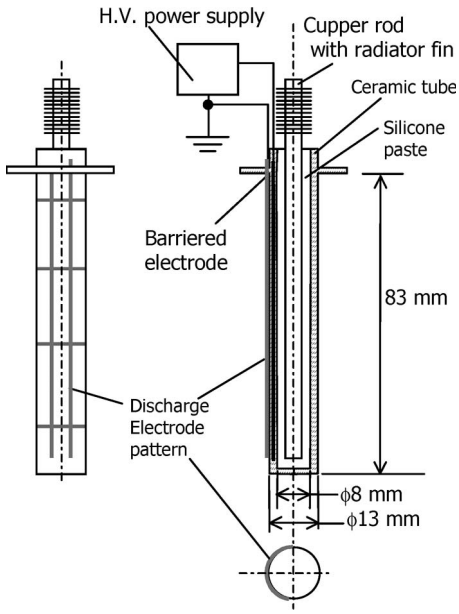


Fig. 3. Surface discharge element for NTP reactor.

The plasma reactor unit was built based on previous experimental results using 12 independent surface discharge elements (OC-002, Masuda Research Inc.). A schematic of the element is shown in Fig. 3. A 1-mm active area was considered around each element. The discharge power that each element can accept according to technical specifications is 10 W as the average with 20 W as the peak value. The heating associated with higher discharge power can be at these values, successfully dissipated using the built-in radiator. The versatile HV supply (HC-RV Masuda Research) with independent voltage and current limited output performs 10 kVpp, 10 kHz, and max 5 A.

Flow rate monitoring through the process prevents gas leak. Discharge power was computed on a digital oscilloscope (Yokogawa DL1740, 1GHz) from the current probe's (Tektronix P6021 current probe) and HV probe's (IWATSU HV-P60) output signals.

The NO concentration of the input gas and the $NO_x(= NO + NO_2)$ amount in the output gas were measured with the gas analyzer (Horiba PG-235: Chemiluminescence NO-$NO_2$-$NO_x$).

Each of the study objectives, namely, flow rate versus concentration, number of discharge elements, distance between elements, and their active region, has been defined aiming at accurate results and process understanding. All experiments have been performed in controlled environmental conditions (21 °C, 45% relative humidity).

### A. Flow Rate Versus Concentration

The amount of nitrogen oxides that can be stored in the absorbent is limited; therefore, desorption process will require the decomposition of NO diluted at various rates in nitrogen. A study of how this various concentrations affect efficiency

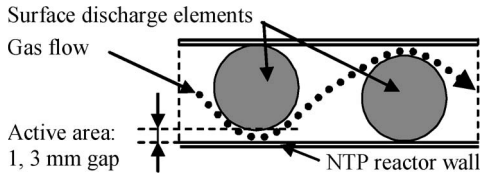


Fig. 4. Testing active area setup for surface discharge elements.

is required. In this setup, the simulated gas was obtained by diluting 2 L/min of 2% balanced NO with $N_2$ at various flow rates (2–20 L/min). The controls of the power supply for voltage and current were set to output top levels delivering maximum power to the surface discharge elements.

### *B. Number of Surface Discharge Elements*

The number of surface discharge elements obviously influences reactor size and cost. In this experiment, the number of surface discharge elements varied from eight to 12. Each of the cases, namely, the energy efficiency, reduction efficiency, and discharge power per element, was determined in four points defined by the total supplied discharge powers: 42, 80, 120, and 160 W. The flow rate and NO concentration were kept constant in all cases at 21.19 L/min and 2560 ppm, respectively.

### *C. Distance Between Surface Discharge Elements*

Overheating of the electrodes is a major concern particularly when lower gas flow rates are used. First reactor models used 30 and 40 mm as the distance between plasma electrodes. In these reactors, at low flow rates ($<$ 10 L/min), the temperature rose significantly after 30 min, threatening the structural integrity of the last electrodes in the row. The reactor used in this experiment maintains a distance of 70 mm between the 12 electrodes ensuring safe operating conditions. However, we wanted to know if further increasing the distance between elements could have an influence on NO reduction efficiency. Using either the first six elements, the six odd elements, the six even elements, or the last six elements, we compared, for a flow rate of 21.9 L/min and NO concentration of 2560 ppm, the average measured values of reduction efficiency for 42-, 80-, and 120-W discharge power values.

### *D. Active Region of Surface Discharge Elements*

For the surface discharge elements to be effective, it is essential for gas flow to pass through a close vicinity (the plasma active area) of their surface. However, as we reduce the gap, the flow speed is increasing together with the pressure drop. Using the setup shown in Fig. 4, we wanted to know the effect of 1-, 2-, and 3-mm gas flow sections on the energy efficiency and NO reduction rate.

A reactor with three elements was used for this experiment while keeping a full 12-electrodes load on the power supply. Flow rate and NO concentration were set at 20.5 L/min and 2110 ppm. Reduction rate was measured in four points at 42-, 80-, 120-, and 160-W discharge power levels.

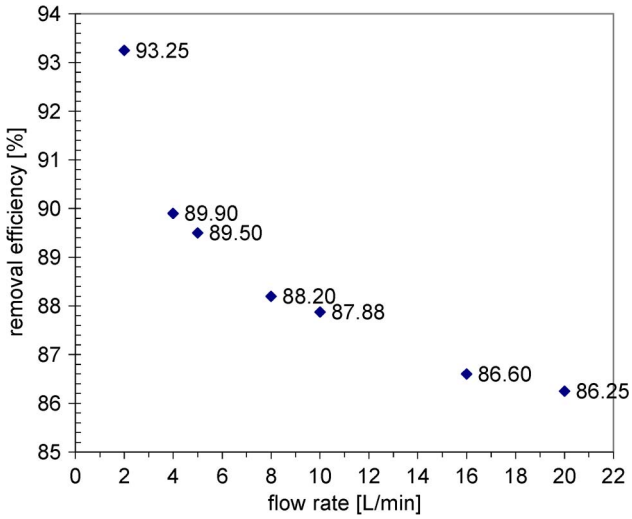


Fig. 5. Reduction efficiency of NO 0.04 [L/min] diluted in $N_2$ at various flow rates (average values of five measurements).

## III. Experimental Results and Discussion

In order to reduce the overall experimental error, successive experiments conducted in similar conditions were compared with results obtained at different moments, disturbance factors being identified and their influence analyzed. We found that overheating of discharge elements and variations of pressure in the simulated gas tubes can lead to significant differences in experimental results.

### *A. Flow Rate Versus Concentration*

Fig. 5 shows the variation of NO reduction efficiency [%] as a function of the flow rate. The energy efficiency was almost constant $29.5 \sim 30.1$ g$(NO_2)$/kWh. The reduction efficiency is higher at low flow rate and higher concentration. This can be partially explained by the longer residence time of gas in the plasma reactor. Another explanation resides in the more efficient use of the energy transferred to gas molecules when NO concentration is higher. A logarithmic decrease of measured discharge power as flow rate increases in Fig. 6 can be linked to the cooling effect of gas flow at higher rates and a reactor capacitance change. The outer temperature of the plasma reactor after half hour of continuous functioning at maximum discharge power did not overpass 65 °C at 2 L/min, and a 4 °C temperature decrease was measured after the flow rate was set to 20 L/min.

### *B. Number of Surface Discharge Elements*

The reduction efficiency and reduction rate showed a strong linear dependency on applied discharge power, but the variation with the number of electrodes was within experimental error limits as can be observed in Table I. The increased plasma volume due to higher discharge power/electrode ratios overcompensates the reduced number of electrodes. This result must be analyzed with dissipated power per element in mind. While the top efficiency for the eight elements is achieved at 20 W/element, with extensive heat generation, the same effect

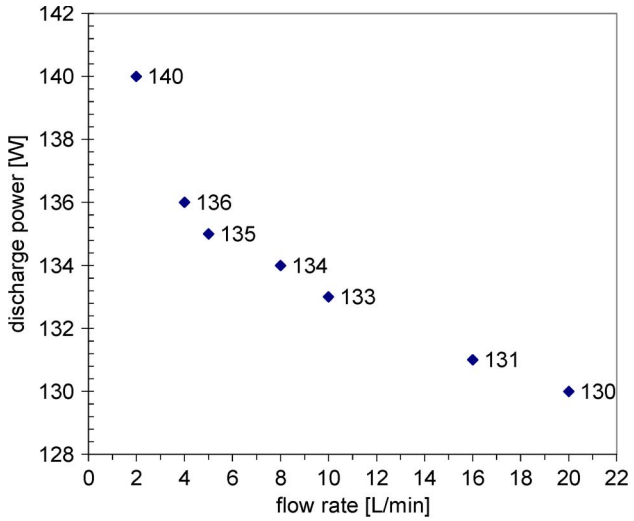


Fig. 6. Influence of gas flow rate on discharge power (each point is the average value of five measurements).

TABLE I
ELEMENTS VERSUS ENERGY AND REDUCTION EFFICIENCY

| Discharge Power [W] | | Reduction efficiency [%] | | | Energy efficiency [g($NO_2$)/kWh] | | |
|---|---|---|---|---|---|---|---|
| | Elements | *12* | *10* | *8* | *12* | *10* | *8* |
| | *42* | 30.5 | 29.7 | 29.7 | 44.4 | 43.3 | 43.3 |
| | *80* | 47.5 | 47.3 | 48.4 | 36.3 | 36.2 | 37.0 |
| | *120* | 64.5 | 64.8 | 66.4 | 32.9 | 33.1 | 33.9 |
| | *160* | 79.2 | 79.9 | 80.1 | 30.3 | 30.6 | 30.6 |

can be achieved at 13.3 W/element in the case of 12 with significant less structural impact and heat generation. The robustness of the system has a high practical benefit: Electrode failure can be compensated in industrial applications if the discharge power is maintained.

### C. Distance Between Surface Discharge Elements

When deciding the distance between elements, several effects must be considered, like heat dissipation, reaction zone, and pressure drop, and not, at least, the dimensional impact on industrial application. The results presented in Fig. 7 show little impact (comparable to experimental error) on reduction and energy efficiency when distance between elements is increased from $x = 70$ mm to $2x = 140$ mm. Measurements on the electrode radiator and external reactor surface were conducted in order to determine equilibrium temperatures. If the distance between the electrodes was reduced to 40 mm or less, the electrode temperature was over the recommended levels, and structural damage was observed.

### D. Active Region of Surface Discharge Elements

Determining the active region of the NTP elements is a key factor for the successful reactor design. In the first development phase, distances as far as 3 mm were considered with pressure drop at high flow rates in mind. Determining optimum operation conditions is a tradeoff between a linear decrease in energy efficiency and an increase in reduction efficiency with discharge power. However, as the results show in Fig. 8, a 1-mm gap presents both higher energy efficiency and higher reduction efficiency for any discharge power value in the given range.

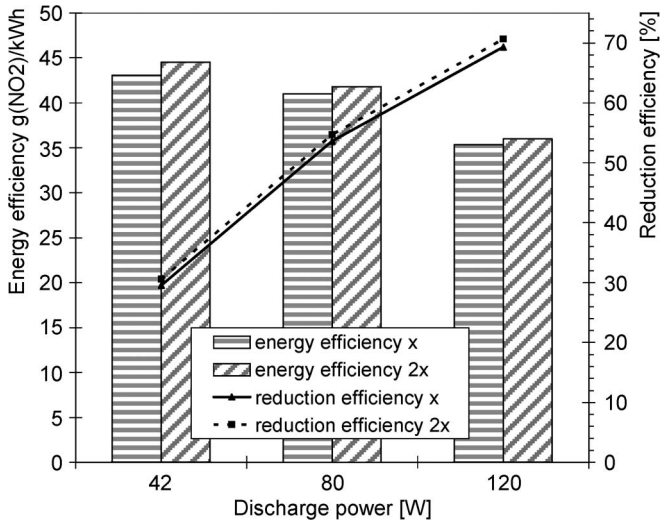


Fig. 7. Comparison of energy and reduction efficiency for a distance between surface discharge elements of $x = 70$ mm and $2x = 140$ mm.

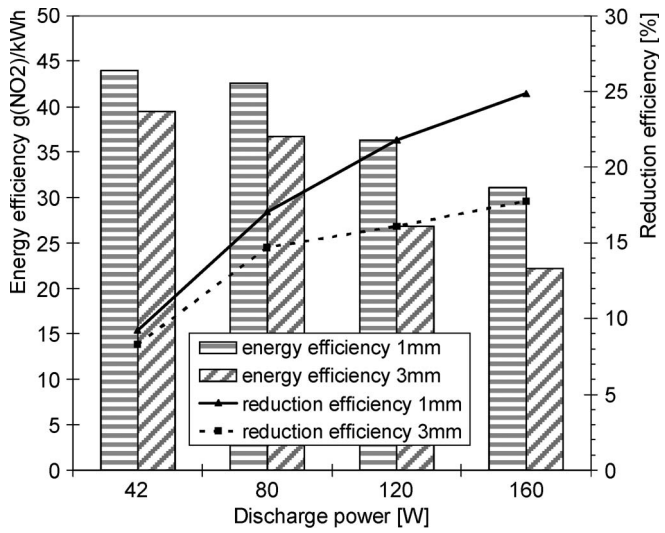


Fig. 8. Comparison of energy and reduction efficiency for 1- and 3-mm gap.

## IV. CONCLUSION

In the "Plasma Combined Exhaust Gas Aftertreatment System for Super Clean Diesel Hybrid Engine," an 86% NO reduction efficiency from a 2000-ppm NO concentration can be achieved after the thermal desorption process with 30 g($NO_2$)/kWh energy efficiency, a performance qualifying the NTP reactor for industrial application. Reduction efficiency decreases with NO dilution. For constant discharge power and flow rate, energy efficiency increases with concentration while reduction efficiency decreases. This observation is stating

the output requirements of a thermal desorption process. The number of NTP elements has little influence on NO reduction as long as the discharge power remains constant. However, because of load specifications, a small number will limit the safe power range, thus, limiting reduction efficiency. Increasing the distance between elements over 70 mm has no visible effect on reduction efficiency. If the active area around the elements is limited to 1 mm, the performance significantly increases compared to a 3-mm configuration. A robust and flexible $NO_x$ reduction in nitrogen NTP system was demonstrated.

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