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Micro-discharge in porous ceramics for exhaust gas cleaning

Jun Sawada*, Yoshihiko Matsui*, Karol Hensel**, Ippei Koyamoto*, Kazunori Takashima*, Shinji Katsura* and Akira Mizuno*

* Department of Ecological Engineering, Toyohashi University of Technology
Tempaku-cho, Toyohashi, Japan 441-8580

** Comenius University, Faculty of Mathematic, Physics and Informatics
Department of plasma Physics, 842-48 Mlynska dolina, Bratislava, Slovakia

In this study, a novel NO_x removal method using micro-discharge in porous ceramics for exhaust gas cleaning was examined. Micro-discharge consists of many discharges in narrow channels inside porous ceramics, and transition to flashover can be suppressed due to narrow channel. The micro-discharge was generated by 60Hz ac high voltage using 29 mm diameter ceramics plate with 10 W electrical input, and was applied for the cleaning of exhaust gas from diesel engine. Two kinds of ceramics with different pore size of 15 μ m and 90 μ m were used. Simulated gas consisted of NO, O₂, N₂ (0.01%, 10%, balance), and C₂H₄ (0%, 0.3% or 0.6%) was supplemented as additive at some conditions. NO oxidation experiments were carried out at 20°C and 150°C. As a result, oxidation ability without the C₂H₄ addition was higher than that with the additive at 20°C.

INTRODUCTION

Nitrogen oxides (NO_x) in exhaust gas from internal combustion engines, especially from diesel engines, cause air pollution and health problems. Conventional NO_x removal process for automobile engines is a catalytic process. Such a catalytic process, however, requires high temperature and low oxygen concentration. Recently, non-thermal discharge plasma or combination of non-thermal discharge plasma and catalysts has been extensively investigated for NO_x removal, because non-thermal discharge plasma can induce chemical reactions under moderate temperatures [1]. The packed bed type is a NO_x removal reactor that can be used in combination with pellet type catalysts. In this reactor, an intense plasma is generated in limited area around many contact points between the pellets. The packed bed reactors achieved good performance in spite of the small plasma volume [2]. However, most catalysts used in practical processes are honey-comb structure, because it has high conversion efficiency, a large surface area and prominent durability. Therefore, combination of plasma and honey-comb process should be very effective for NO_x treatments. If an effective combination of plasma and honey-comb catalyst is developed, the performance will be improved.

For this purpose, we examined the NO_x removal using the micro-discharge. Porous ceramics were used to generate the micro-discharge due to electrical breakdown in micro channels. This micro-discharge can possibly be used to generate discharge plasma in honey-comb catalyst having porous structure with microchannels.

MICRODISCHARGE IN POROUS CERAMIC PLATE

When an Electrostatic Precipitator (ESP) is used for dusts with apparent resistivity more than about $5 \times 10^{10} \Omega \text{ cm}$, breakdown takes place in a dust layer. This is called back corona. In this research, we examined

a NO_x removal process using back corona, or electrical breakdown inside the ceramics. This process can be used for the combination of a catalyst and non-thermal discharge plasma.

Fig.1 shows a schematic illustration of the micro-discharge in porous ceramics plate. AC or DC high voltage is applied to a needle (or a mesh electrode placed on the surface of the ceramics instead of the corona electrode), and corona discharge charges up the surface of the ceramics to cause breakdown (1, 2) The breakdown can be confined in narrow channels of the ceramics, and transition to arcing can be avoided.(3), and this microdischarge can be stabilized.

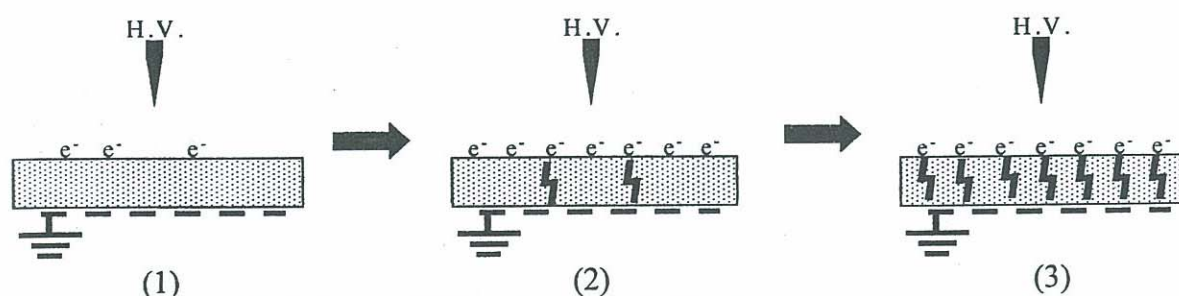


Fig.1 Schematic illustration of micro-discharge (e^- : negative ionic charge)

EXPERIMENTAL SETUP

Fig.2 shows a micro-discharge reactor. Experiments were carried out using AC 60 Hz high voltage. Two disk electrodes (18 mm-diameter) made of fine stainless steel mesh were placed on both sides of the ceramics disk. Disk-type porous ceramics plates (3 mm thickness and 29 mm diameter) were used in the experiment. Table.1 shows the properties of the ceramics. Ceramics A and B with different pore size were sintered from alumina powders of different size.

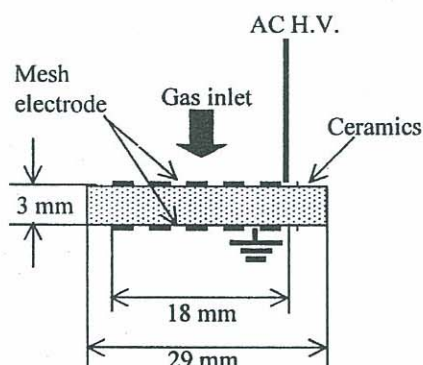


Table.1 Properties of the ceramics

	Al ₂ O ₃ contents [%]	Pore size [μm]	Powder size [μm]	Porosity [%]
Ceramics A	92	15	40	42
Ceramics B	92	90	180	37

Fig.2 Reactor of micro-discharge

Fig.3 shows a layout of the experimental setup. The gas composition of NO, O₂, C₂H₄ and N₂ was adjusted using mass flow controllers. The simulated gas contained 100ppm NO, 10% of oxygen and 0 or 300 ppm C₂H₄ in N₂ base gas. Gas flow rate was 0.6 LPM (liter per minutes) and the pressure loss were 1.5 kPa and 140 Pa for ceramics A and B, respectively. The input power was kept constant at 10W in all experiments.

The reactor was placed in an oven, and the temperature was set at 20°C or 150°C. The NO and NO_x (=NO+NO₂) concentration were monitored with a NO_x meter (HORIBA PG-250). The concentration of C₂H₄ was measured using GC-FID (Shimadzu GC-14B). The voltage waveform was monitored by an oscilloscope (Tektronix TDS 350) with a high voltage probe (Tektronix P6015) during the experiment.

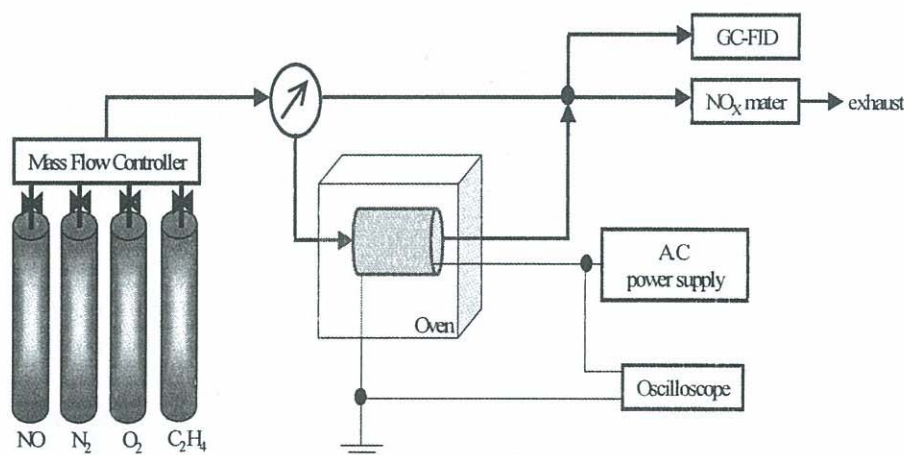
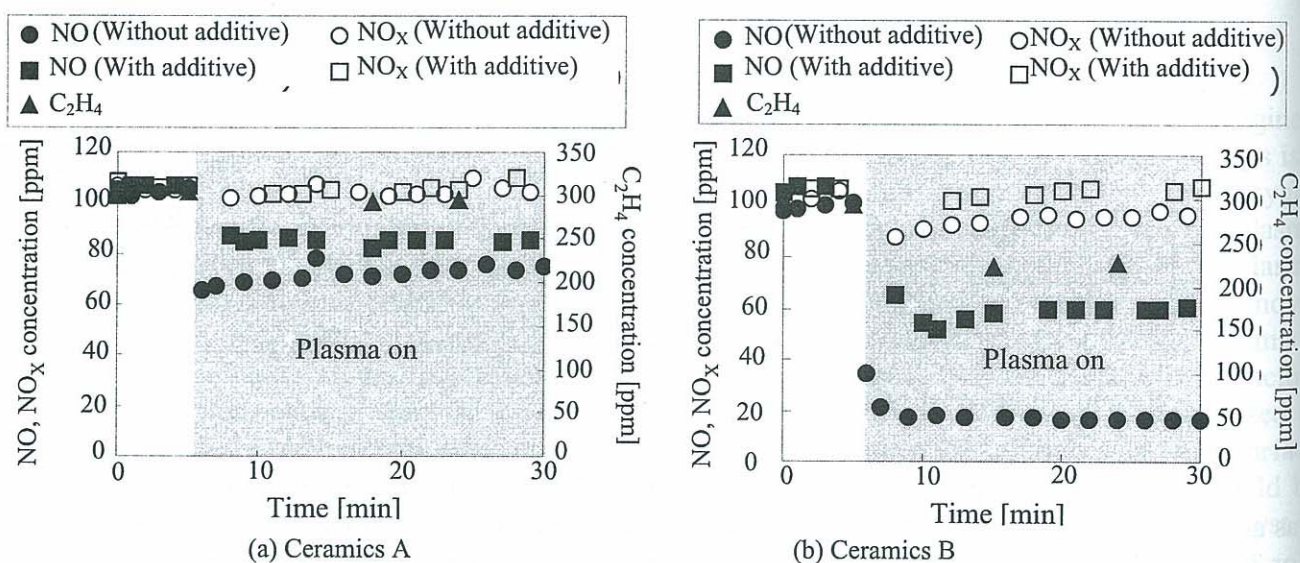


Fig.3 Schematic diagram of the experimental setup

RESULTS AND DISCUSSION

Fig.4 shows the concentration of NO and NO_x with and without C₂H₄ at 20°C. Fig.4(a) and (b) correspond to the results with ceramics A and B, respectively. In Fig.4(a), NO concentration rapidly decreased after starting the voltage application with or without the additive. NO_x concentration, on the other hand, was not changed by the voltage application. These results indicate that NO was oxidized to NO₂ due to the micro-discharge but no further reaction, nor absorption took place to remove NO₂. NO oxidation ability without additive was higher than that with the additive. C₂H₄ was not decreased even when the plasma was generated in this case.

Fig.4 Concentration of NO, NO_x and C₂H₄ at 20°C

-Condition of Fig.4 a, b; Gas flow rate:0.6LPM, Temperature:20°C, Space Velocity, a:38000h⁻¹, b:43000h⁻¹, 300ppm of C₂H₄ was used as additive, AC high voltage(60Hz, a:16kV_{p-p}, b:15kV_{p-p}), Input power :10W

In the ceramics B as shown in Fig.4(b), the oxidation ability of NO with or without the additive were higher than that of ceramics A. Concentration of C₂H₄ decreased by 70ppm. The oxidation ability of NO without the additive was higher than that with the additive in this case, too. Under this condition, the additive (C₂H₄) did not enhance the oxidation of NO. This NO-oxidation characteristics using the micro-discharge were quite different from those using DBD (Dielectric Barrier Discharge) or pulsed corona, in which NO oxidation can be enhanced using C₂H₄ [2].

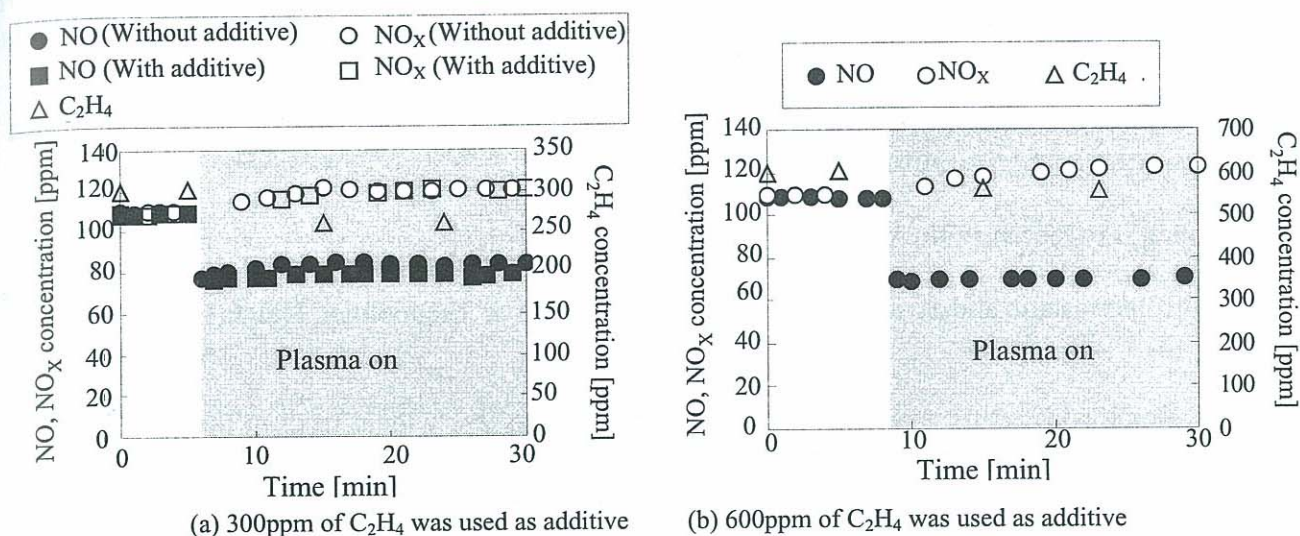


Fig.5 Concentration of NO, NO_x and C₂H₄ at 150°C using ceramics B
-Condition of Fig.5 a,b; Gas flow rate:0.6LPM, AC high voltage (60Hz, 11kV_{p-p}), Input power:10W

The results at 150°C is shown in Fig.5. The experiment was carried out using the ceramic B with (a) 300ppm and (b) 600ppm of C₂H₄ as the additive. In Fig.5(a), the oxidation ability of NO with the additive was slightly higher than that without the additive. The concentration of NO_x with and without the additive were increased by about 10-20ppm when the microdischarge was generated. In Fig.5(b) with 600ppm of C₂H₄, the NO oxidation ability was slightly higher but similar to Fig.5(a).

CONCLUSIONS

We studied the NO_x removal using the micro-discharge. Microdischarge can be generated with ac 60 Hz energization of 10 W input power, using two kinds of ceramics plate of 29 mm diameter of average pore size of 90 μm and 15 μm. The oxidation of NO using ceramics with average pore size of 90 μm at 20°C without C₂H₄ additive showed the highest performance among the conditions tested in this study.

REFERENCE

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- [2] Yoshihiko Matsui et al., SAE Technical paper, No.2001-01-3511