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exist for the publication of theoretical and experimental investigations of all aspects of the mechanics and thermodynamics of fluid-flow with special reference to fluid-flow machines

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PRACE INSTYTUTU MASZYN PRZEPŁYWOWYCH

poświęcone są publikacjom naukowym z zakresu teorii i badań doświadczalnych w dziedzinie mechaniki i termodynamiki przepływów, ze szczególnym uwzględnieniem problematyki maszyn przepływowych

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NO_x treatment test by positive DC corona reactor with and without water

We propose a new type of corona reactor for NO_x treatment in fuel burned exhaust gas. The reactor operates with positive DC corona discharge on saw-edge electrode over water surface. The positive DC corona produces effective radicals for NO oxidization and helps NO_2 dissolving into water with corona wind blowing down to water surface. First we show the positive and negative DC corona characteristics over water surface with multi needle and saw edge electrodes. Then we show some typical experimental results of NO_x treatment tests with diesel exhaust gas and dry NO test gas. The experiments were carried out under some different discharge modes and inlet gas concentrations to find the best operative conditions of this reactor and to investigate the possible radicals or reactions as the effect of water existence. We got more than 95% removal rate of NO_x with 200 ppm dry gas by positive corona with water. In case of diesel exhaust gas, more than 90% of NO_x removal could be achieved with and without water. The results of our experiments suggest that existence of water molecules (as vapor or fume) and OH radicals in the reactor are important for performing the effective reactions of NO_x removal processes.

1. Introduction

Study on NO_x treatment technology by nonthermal plasma is developing with recent experimental and theoretical researches [1,2]. Some effective radicals and plasma chemical reactions contributing to NO_x removal processes have been listed up with their chemical reaction energy by Dr. Chang et al. [3]. Recent studies suggest that humidity or OH radical in discharge space should be important to promote the reaction for NO_x treatment processes.

Mizuno group carried out NO_x removal test and analyzed the reaction processes on dry and water film type reactor by pulsed corona [4]. Dors and Mizeraczyk group also reported on NO_x removal tests with pulsed wet type electrostatic precipitator model [5]. They got remarkable results in NO_x removal and discussed on

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some plasma chemical reactions promoted by existence of water. And they also discussed on possibilities and problems for industrial application.

On the basis of the same idea, we propose an advanced NO_x treatment reactor with positive DC corona over water surface. We have carried out NO_x treatment tests under various conditions in different discharge electrodes, with and without water, as well as test gas concentration etc. The DC corona reactor is more convenient in practical application than pulse corona reactor due to its lower cost performance for preparing the power supply.

Generally, most of NO_x in exhaust gas exists initially as NO, which is low water-soluble. NO_x removal processes in this reactor should be expected that ozone and radicals produced over water surface by corona discharge promote oxidation of NO to NO_2, which subsequently dissolves into water effectively with a help of corona wind toward water surface [6].

To confirm the reaction in the reactor, NO_x treatment tests were carried out on positive and negative DC corona to water surface or to plate electrode without water. Both diesel engine exhaust gas and NO mixture dry gas were prepared for the test gas.

In this paper, we show some suggestive results that will help us to understand the effective chemical reactions in NO_x removal processes.

2. Experimental setup

The corona reactor for test is hand made rectangular prism box as shown in Fig. 1. Upper discharge electrode is steel plate with 14 needles or 4 saw edge of 10 cm length. Lower electrode is a stainless steel plate placed in water or a metal rod soaked 10 mm of its top in water. These electrodes are arranged inside of a sealed plastic box of the size $10 \times 5 \times 20$ cm. Distance from needle top or saw edge to the water surface is to be "a", to plate electrode in water to be "b" and depth of water to be "c".

Discharge characteristics of each type of reactors were measured under various conditions before NO_x treatment test. Test gases from diesel engine flew into the reactor with 5.0 l/min of flow rate and passed through two stage commutators into discharge reaction section. NO_x and NO concentrations at the outlet of reactor were measured separately by NO_x analyzer changing the corona discharge currents up to the spark voltage. To confirm the effect of this reactor as an electrostatic precipitator, we measured simultaneously dust concentrations in output gases by a laser particle counter in case of diesel exhaust gas test.

Pure NO gases blended with clean and dry air as some different concentrations were also used with the flow rate of 5.0 l/min. We call it a dry gas or blended gas tests. In case of pure water reactor, pH and conductivity of reactor's water were measured before and after the gas treatment test. All experiments were carried out under normal room temperature and pressure.

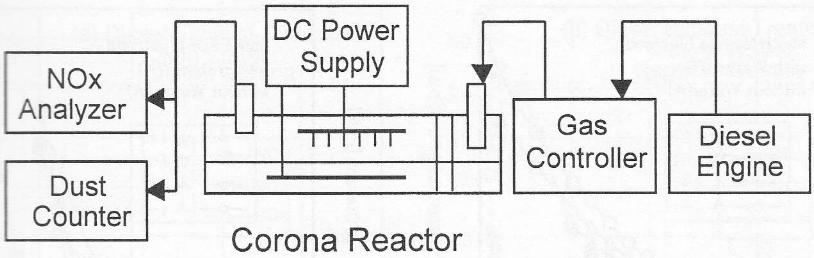

Fig. 1. Experimental layout.

Table 1. Experimental conditions

Reactor:
Size: 50 × 100 × 200 mm
Discharge Electrode: Multi Needle, Saw Edge
to Flat Plate (without Water), to Water Surface(with Water)
Discharge Mode:
DC, Positive or Negative Corona
Test Gas:
Diesel Engine Exhaust (NO_x Concentration 10, 20, 60 ppm)
Blended Dry Gas (Air + NO, NO_x Concentration 20, 60, 200 ppm)
Gas Flow Rate:
6.0 l/min (2.4 cm/sec)
Measurement:
NO_x Analyzer; Horiba CLA51S, NO, NO_x
Dust Counter; Shibata LD1, 0.6 μm
Ion Analyzer; Orion Model 720A, NO_{2-}, NO_{3-}

3. Results

3.1. Corona discharge over water surface

Fig. 2 shows positive and negative corona characteristics of multi needle electrode to water surface or to plate electrode with a = 10 mm. Corona currents depend on distance of space between the needle top and the water surface "a". Locations of lower electrode in water and conductivity of water do not give much influence in V-I characteristics.

With the increase of the applied voltage, corona discharge starts at the needles' top and extends toward water surface. Stabilized corona could be maintained up to the spark occurrence voltage. Corona wind down to water surface occurs with a start of the corona discharge and it makes caves and waves on the water surface.

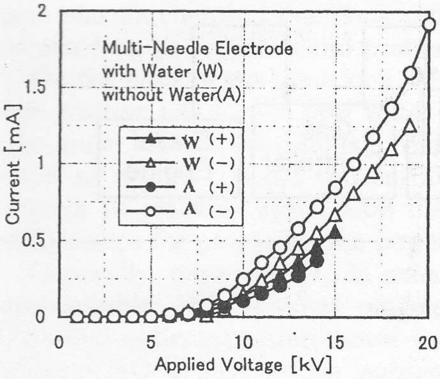

Fig. 2. Discharge characteristics of reactor of multi needles electrode with water (W) and without water (A).

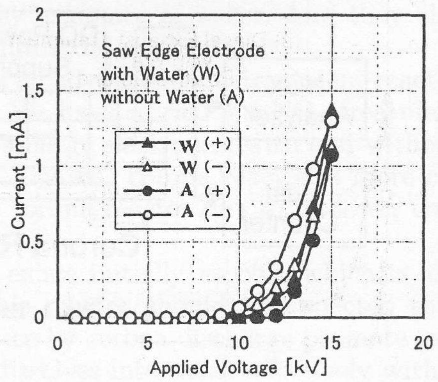

Fig. 3. Discharge characteristics of reactor with saw edge electrode with water (W) and without water (A).

Positive and negative corona currents are shown in Fig. 3 measured by a saw-edge electrode for an upper electrode placed as parallel to water surface or lower plate electrode. With saw-edge electrode, discharge current and spark voltage are not so much changed by its polarity of applied voltage. This fact means we can gain high corona current and energy with saw-edge upper electrode even in positive without spark. In practical reactor with water, the lower plate electrode can be replaced by a metal rod electrode soaked into water of its top.

3.2. NO_x treatment tests

All results shown here on NO_x treatment tests were obtained by the experiments with positive and negative discharge with saw edge electrode as the discharge electrode. Fig. 4 shows NO_x concentrations and the removal rate versus discharge current with the diesel exhaust gas. Inlet gas into the reactor was controlled as 50-60 ppm of NO_x concentration from the diesel engine. Reactor with water (a, c) shows good removal rate of NO and NO_x with the increase of the discharge current. In the case of a reactor without water (b, d), the removal of NO is almost the same as with water reactor and NO_x removal rate is a little lower.

Fig. 5 shows results of NO_x treatment tests with dry NO gas when input NO_x concentration was controlled to remain at the level of about 200 ppm. In this figure, outlet NO, NO_2 and NO_x concentrations vs. discharge current are shown in the reactor with water in positive corona operation (a), and without water in negative corona operation mode (b).

Characteristics of treatment by the reactor with water demonstrate that NO rapidly decreases with the increase of the corona current. Similarly the concentration of NO_x also decreases with the increase of corona current by the reactor

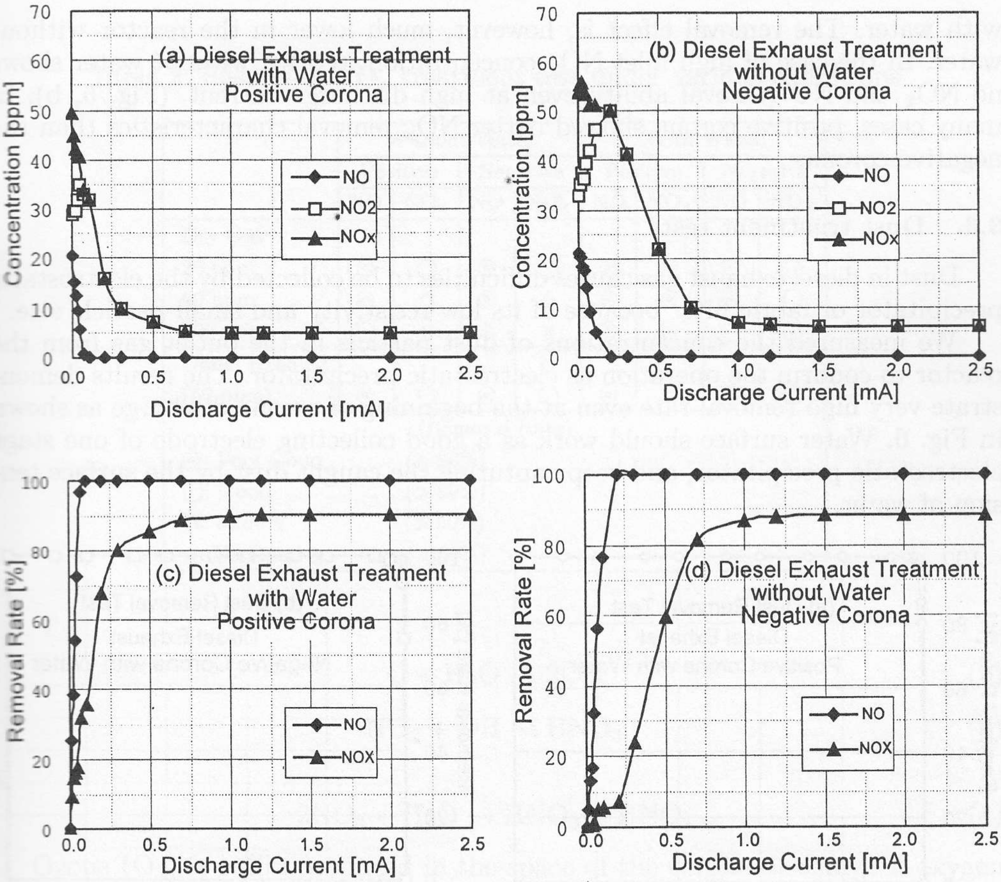

Fig. 4. NO_x concentration (a,b) and removal rate (c,d)vs. discharge current by the reactor with water (a,c), and without water (b,d) (Diesel Exhaust Gas).

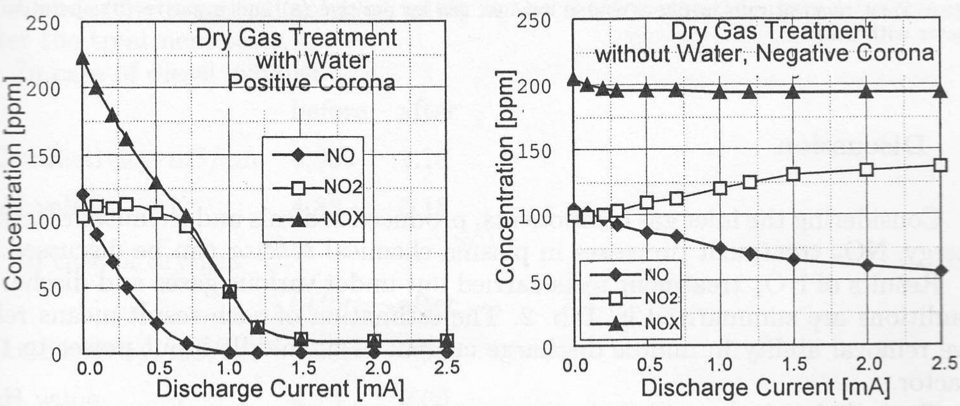

Fig. 5. NO_x concentrations by water (a) and without water (b) when inlet dry gas of 200 ppm.

with water. The removal effect is, however, much lower in the reactor without water. In the case of high inlet NO_x concentration, reactor without water shows no NO_x and NO removal ability even at high discharge current. (Fig. 5, b). In many cases, positive corona showed better NO_x removal characteristics than the negative corona.

3.3. Dust treatment tests

Dust in diesel exhaust gas causes difficulties to be collected by the electrostatic precipitator or fabric filter because of its low resistivity and small particle size.

We measured the concentrations of dust particle in the outlet gas from the reactor to confirm the operation as electrostatic precipitator. The results demonstrate very high removal rate even at the beginning of corona discharge as shown in Fig. 6. Water surface should work as a good collecting electrode of one stage electrostatic precipitator, and keep capturing the caught dust by the surface tension of water.

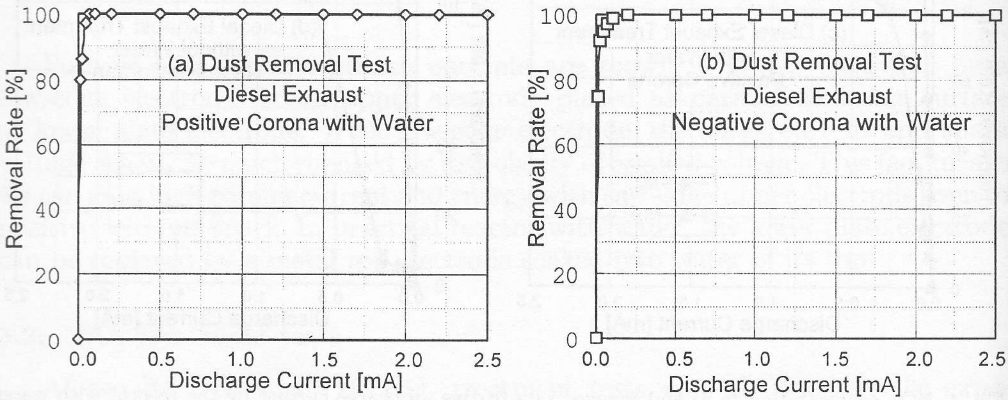

Fig. 6. Dust removal rate in diesel engine exhaust gas by positive (a) and negative (b) operation of reactor with water.

4. Discussion

Considering the inlet gas components, produced radicals and chemical reaction energy, NO_x treatment processes in plasma chemical reactor can be discussed.

Results of NO_x treatment tests carried out under various gases and discharge conditions are summarized in Tab. 2. The estimation of each result means relative removal ability in middle discharge current or at middle input power to the reactor.

Typical chemical reactions in the reactor should be expected as,

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (1)$$

Table 2. Results of NO_x treatment test under various conditions

	Without Water				With Water			
	Positive		Negative		Positive		Negative	
	NO	NO_x	NO	NO_x	NO	NO_x	NO	NO_x
Dry Gas								
20 ppm	⊚	○	⊚	○	⊚	⊚	⊚	○
60 ppm	⊚	△	⊚	△	⊚	⊚	⊚	○
200 ppm	○	×	△	×	⊚	⊚	○	○
Diesel Exhaust (60 ppm)	⊚	○	⊚	○	⊚	⊚	⊚	○

(Removal Rate)

⊚: Very Good (> 90%)
○: Good (> 80%)
△: Middle (> 50%)
×: No Good (< 50%)

$$O + H_2O \rightarrow 2OH \tag{2}$$

$$NO_2 + OH \rightarrow HNO_3 \tag{3}$$

or

$$2NO_2 + H_2O \rightarrow HNO_3 + HNO_2 \tag{4}$$

Ozone (O_3) is easily produced in the space of the corona discharge if oxygen contents (O_2) reaches more than few percent of concentration. In the reactor with water, humidity is supplied to the discharge space and helps enhancing these reactions. In addition, the corona wind helps NO_2 to dissolve into water.

Conductivity and pH value of water in the reactor were measured before and after the treatment tests.

In case of diesel exhaust

	before	after
Conductivity, mS/cm	0.009	1.18
pH value	4.83	3.18

In case of dry gas

	before	after
Conductivity, mS/cm	0.009	1.96
pH value	4.83	2.06

We confirmed the increase of NO_3 and NO_2 ions in water of the reactor after

30 min of dry gas treatment test. With these background frames, the typical results of our experiments should be explained.

In case of treatment of dry gas as shown in Fig. 5(b), the lack of humidity or OH radical may cause the low NO_x removal rate, which should be on the other hand, induced by reaction (2) and (3).

In diesel engine exhaust gas, humidity and complex small dust particles should give effect of absorption of NO_2 or NO_3 even with the reactor without water. After that they should be collected on a plate electrode or on a filter at the entrance of NO_x analyzer.

5. Conclusion

The corona discharge over the water surface shows following remarks:

(1) Stable positive and negative DC corona discharge could be obtained between multi-needles or saw-edge and water surface.

(2) The discharge demonstrated some benefits for application to design the reactor for NO_x and dust treatment of combustion gases.

Results of NO_x treatment tests by the reactor with water demonstrate following remarks:

(3) In the case of diesel exhaust gas, NO_x removal rate showed to be better in a reactor with water rather than that without water.

(4) For blended dry gas, the reactor with water showed significant NO_x removal effect especially in the case of high NO concentration.

In the case when this reactor operates as an electrostatic precipitator,

(5) Very high collecting rate of dust particle could be achieved as well as NO_x removal.

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Usuwanie NO_x w reaktorze stałonapięciowego wyładowania koronowego z wodą i bez wody

Streszczenie

Autorzy pracy zaproponowali nowy typ reaktora do usuwania NO_x z gazów spalinowych, w którym dodatnie stałonapięciowe wyładowanie koronowe generowane było pomiędzy elektrodą o ostrzu piły a powierzchnią wody. W wyładowaniu powstawały rodniki utleniające NO do NO_2 wspomagające proces rozpuszczania NO_2 w wodzie. W pierwszym etapie prac określono charakterystykę prądowo-napięciową stałonapięciowego wyładowania dodatniego i ujemnego. W etapie drugim działaniu wyładowania poddano gazy spalinowe z silnika Diesla oraz mieszaninę powietrza z NO. W wypadku gazów spalinowych uzyskano ponad 90%-ową eliminację NO_x. Wyniki badań świadczą o istotnym wpływie cząsteczek wody i powstających z nich rodników OH na efektywność eliminacji NO_x.