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NO_X REMOVAL IN A CORONA DISCHARGE - CATALYST HYBRID SYSTEM

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A b s t r a c t. The objective of this work was to investigate NO_x removal employing a hybrid system of a dc corona discharge and a catalyst at room temperature (22°C) in the presence of ammonia. Results of the investigation showed that the hybrid system removes up to 96% of NO_x with an energy efficiency of 3.4 g NO/kWh. Without the catalyst, NO_x decomposition in the "pure" corona discharge with ammonia was lower (up to 66%). Also the energy efficiency was lower (about 1.8 g NO/kWh). The obtained results proved that the investigated hybrid system consisted of the corona discharge and the catalyst is attractive for NO_x removal because relatively high efficiency both in decomposition and energy.

K e y w o r d s: corona discharge, catalyst, nitrogen oxides.

INTRODUCTION

Ten-old years investigations carried out in laboratories and pilot plants showed that removal of NO_x from flue gases by corona discharges may be very efficient [1, 2]. However, the energy consumption of this new technology is still not acceptable for commercial use and is to be improved. Thus, many recent investigations have been focused on the performance improvement of the corona discharge process by optimising the power source, using gaseous additives or combining with other methods.

A competitive technology the *selective catalytic reduction (SCR) process* has proved its efficiency in NO_x removal in both selectivity and high performance [3]. In this process, ammonia and a catalyst operating in a relatively high temperature range of 280-430°C are used to reduce NO_x to N_2 that is harmless product. In the *corona disccharge process* the dominant mechanism of NO_x removal is oxidation to removable HNO_3, and the selective control of the plasma chemical reaction to reduce NO_x to N_2, like in the SCR, is difficult. However, in recent years hybrid plasma systems have been proposed because of their capability to decompose NO_x

into N_2 and O_2 [4-9]. A typical hybrid system consists of a corona discharge and a catalyst. So far researchers used a catalyst either as a layer coating the plane electrode in a point-to-plane DC corona discharge reactor [4], or as pellets or globules in a packed bed silent discharge reactors [5-9]. In some hybrid systems, hydrocarbons were added to the air or flue gas polluted with NO_x to improve its removal [5, 6, 8]. In the others, ammonia was used as an additive after the corona processing, before the SCR processing [7, 10-12].

The objective of this work was to study NO_x removal by a combined system of a DC streamer corona discharge with a catalyst under low temperature (22°C) in the presence of ammonia. In this system ammonia is added to the gas before entering corona discharge reactor.

EXPERIMENTAL APPARATUS

The non-thermal plasma reactor with the corona discharge and catalyst used in this experiment is shown in Fig. 1. The positive DC corona discharge was generated between a stainless steel hollow needle and a flat mesh (1x1 mm) made of brass. The needle was placed perpendicularly to the mesh. The outer and inner diameter of the needle was 2 mm and 1.6 mm, respectively. The interelectrode distance was varied in the range of 20-40 mm. Gas mixture of N_2(80%):O_2(5%):CO_2(15%):NH_3(250ppm):NO(200ppm) flowed through the hollow needle with a flow rate of 1 l/min. The catalyst, typical of the SCR processing, used in the investigation was a mixture of V_2O_5 and TiO_2 deposited on the Al_2O_3 globules of 5-6 mm in diameter. The globules with deposited catalyst were delivered by the Katalizator Co., Krakow, Poland. The catalyst globules were placed on the mesh in the form of 2 or 4 layers, so that the interelectrode distance was always about 40 mm.

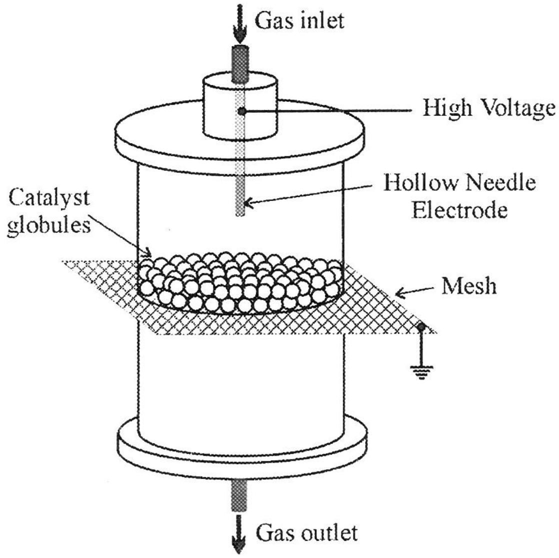


Fig. 1. Non-thermal plasma reactor with corona discharge and catalyst.

The positive polarity DC high voltage was applied through a 10 MΩ resistor to the hollow needle electrode. The operating voltage was varied from 15 to 38 kV to develop a stable streamer corona discharge. The time averaged discharge current was varied in the range of 50-200 μA. Time dependences of the corona discharge current pulses were measured with a current transformer PEARSON 2878 and recorded on a digital oscilloscope.

Concentrations of NO, NO_2 and NH_3 in the gas mixture were measured by absorption spectroscopy method using a Perkin-Elmer 16 PC FTIR spectrophotometer operating in the infrared range 4400-1000 cm^{-1}.

RESULTS

The investigation of NO_x abatement by the corona discharge plasma was carried out either in the reactor without catalyst or in the reactor with 2 layers of catalyst globules. In both cases the interelectrode distance was 40 mm. However, when catalyst is present in the reactor, the distance between the catalyst layer and the needle was about 30 mm.

Generally, the removal of NO and NO_x increases with increasing corona discharge current. In the case of reactor without catalyst, removal of NO from the operating gas not containing NH_3 is no higher than 30 % and is accompanied with NO_2 production (Fig. 2a). As a result the NO_x removal is not higher than 15 %

(Fig. 2b). NH_3 added to the operating gas increases NO removal and completely removes NO_2 produced in the corona discharge plasma. However, only small fraction of the introduced NH_3 is consumed during the corona discharge process. In the presence of the catalyst in the reactor, NO removal from the operating gas without NH_3 is about 50 % and again is accompanied with NO_2 production (Fig. 2a). As a result the NO_x removal does not exceed 35 % (Fig. 2b). In the presence of NH_3, removal of NO and NO_x increases up to 96% with no production of NO_2 and only 2 ppm of NH_3 in the outlet gas.

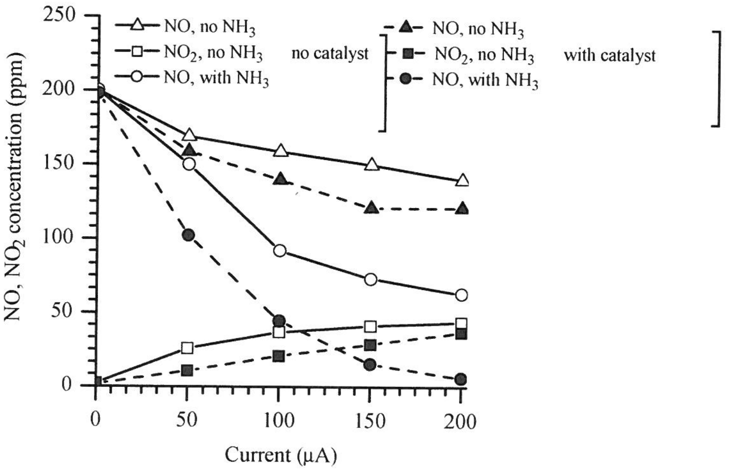
Long time experiment showed that catalyst works stable for about 30 hours (Fig. 3). After that time it loose its activity probably due to covering the globules surface with NH_4NO_3 powder which is formed in the following reaction [10]:

$$2\,NO_2 + 2\,NH_3 \rightarrow N_2 + H_2O + NH_4NO_3. \quad (1)$$

From the weight analysis of the NH_3 and NH_4NO_3 one can conclude that only a fraction of NO_x and NH_3 is transformed into ammonium nitrate. Another reaction responsible for removal of NO_x and NH_3 is reduction of NO_2 [10]:

$$6\,NO_2 + 8\,NH_3 \rightarrow 7\,N_2 + 12\,H_2O. \quad (2)$$

a)



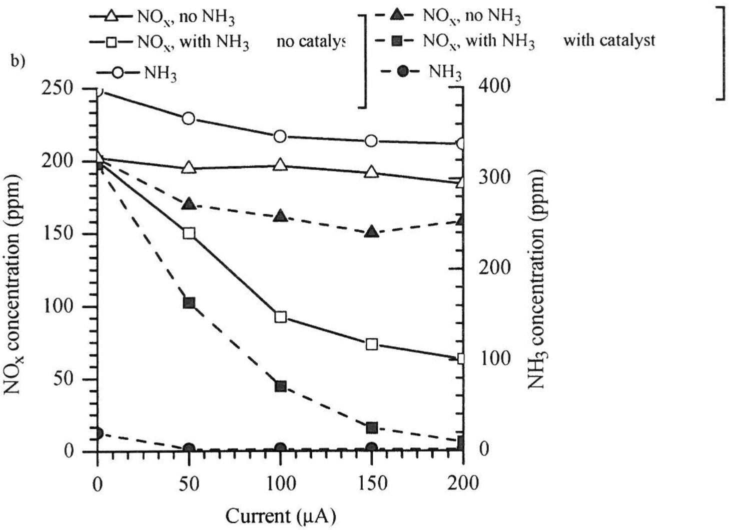


Fig. 2. Concentrations of NO, NO_2, NO_x and NH_3 in the operating gas at the reactor outlet as a function of time-averaged corona discharge current. Operating gas without and with NH_3 (250 ppm). Catalyst globules formed in 2 layers. Interelectrode distance 40 mm.

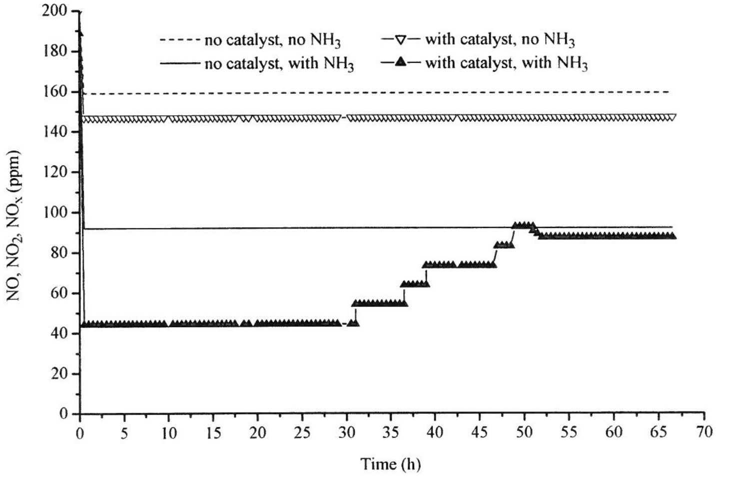


Fig. 3. Time dependence of NO concentration in the operating gas at the reactor outlet. Operating gas without and with NH_3 (250 ppm). Catalyst globules formed in 2 layers. Interelectrode distance 40 mm.

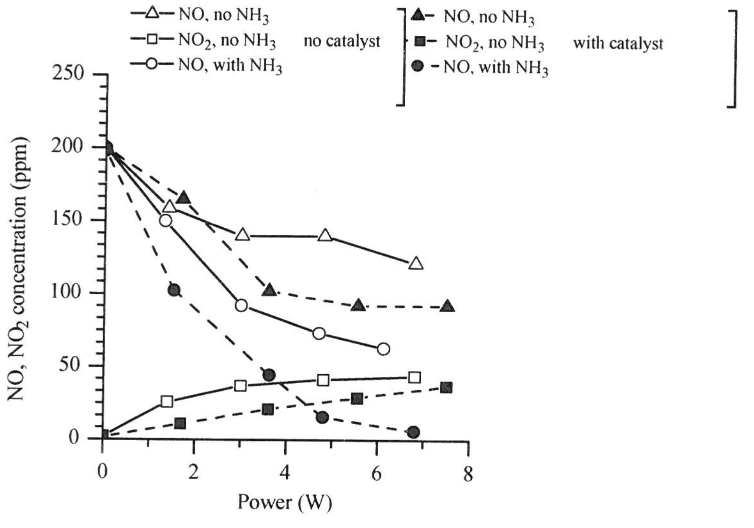


Fig. 4. Concentrations of NO and NO_2 in the operating gas at the reactor outlet as a function of corona discharge power. Operating gas without and with NH_3 (250 ppm). Catalyst globules formed in 2 layers. Interelectrode distance 40 mm.

The energy consumption during NO_x removal process in the corona discharge in the reactor with and without catalyst can be calculated from results presented in Fig. 4.The energy efficiency of the removal of 96% NO in the reactor with catalyst is 3.4 g NO/kWh. This is similar to result obtained by Ohkubo et al. [12] in the corona radical shower reactor with 8 hollow needles. In the case of the reactor without the catalyst (operating gas with NH_3, 400 ppm) the energy efficiency of the NO removal is only 1.8 g NO/kWh.

CONCLUSIONS

The investigation of influence of the catalyst typical for SCR process on NO_x removal from the simulated flue gas in the non-thermal corona discharge reactor showed that the catalyst increases substantially NO_x removal efficiency. Besides, our hybrid system reduces much energy consumption of NO_x removal process what is the main goal of the work. Thus, the obtained results proved that the investigated hybrid system consisted of the corona discharge and the catalyst is attractive for NO_x removal and may be alternative to other non-thermal plasma methods of gas cleaning.

ACKNOWLEDGMENT

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ELIMINACJA NO_X W UKŁADZIE HYBRYDOWYM WYŁADOWANIE KORONOWE – KATALIZATOR

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S t r e s z c z e n i e. Tematem pracy było badanie procesu eliminacji NO_x w układzie hybrydowym DC wyładowanie koronowe – katalizator w temperaturze pokojowej (22 °C) w obecności amoniaku. Uzyskano efektywność usuwania do 96% NO_x przy efektywności energetycznej 3.4 g NO/kWh. W procesie bez katalizatora stopień eliminacji NO_x był niższy (do 66%), jak również efektywność energetyczna (ok. 1.8 g NO/kWh). Otrzymane rezultaty wskazują na atrakcyjność systemu hybrydowego z powodu wysokiego stopnia eliminacji NO_x i wykorzystania energii.

S ł o w a k l u c z o w e : wyładowania koronowe, kataliza, tlenki azotu.