

NO_x removal from a flue gas in a corona discharge-catalyst hybrid system

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Abstract

The objective of this work was to investigate NO_x removal with a hybrid system consisting of a dc corona discharge and a catalyst at a room temperature (22 °C) in the presence of ammonia. The experimental procedure was as follows. A positive dc corona discharge was generated between a stainless steel hollow needle and a flat mesh made of brass. The needle was placed perpendicularly to the mesh maintaining the interelectrode distance of 40 mm. The outer and inner diameters of the hollow needle were 2 and 1.6 mm, respectively. The catalyst used in this investigation was a layer of V₂O₅ and TiO₂ deposited on Al₂O₃ globules of 5–6 mm in diameter. They were placed on the mesh so that the distance between the tube and the globules was 30 mm. The catalyst was either saturated with NH₃ or free of NH₃. A gas mixture of N₂ (80%):O₂ (5%):CO₂ (15%):NO (200 ppm) flowed through the tube with a flow rate of 1 l/min. Ammonia was added to the gas mixture through a hollow needle in a concentration of either 400 ppm when NH₃-free catalyst was used or 30 ppm when catalyst saturated with NH₃ was used. Results of the investigation showed that the hybrid system (with NH₃-saturated catalyst) removes up to 96% of NO_x with an energy efficiency of 3.4 g NO/kWh. Without the catalyst, NO_x removal in the “pure” corona discharge in the gas mixture with ammonia was lower (up to 66%). Also the energy efficiency was lower (about 1.8 g NO/kWh). The obtained results showed that the investigated corona discharge-catalyst hybrid system is attractive for NO_x removal because of relatively high efficiency both in NO_x decomposition and energy consumption.

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1. Introduction

Investigations over the last 10 years 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 in this new technology is still not acceptable for commercial use and is to be improved. Therefore, many recent investigations have been focused on the performance improvement of the corona discharge processing by optimizing the power source [3], using various gaseous additives [4–10] or combining the corona discharge processing with other methods [11–13].

A technology competitive to the corona discharge processing is the selective catalytic reduction (SCR) which has proved its efficiency in NO_x removal [14]. In the SCR process, ammonia (NH₃) and a catalyst operating at a relatively high temperature range of 280–430 °C are used to reduce NO_x to N₂ and H₂O which are harmless products. Although, efficiency of NO_x removal using the SCR method is up to

90%, there are disadvantages of this method, such as high cost and short life-time of the catalyst due to its poisoning.

In the corona discharge processing, the dominant mechanism of NO_x removal is its oxidation to gaseous HNO₃ which then is converted to NH₄NO₃ solid particles by adding NH₃ [5,6,8,9,15]. Among many corona discharge types, the most efficient in NO_x removal is a corona radical shower (CRS) [8,9]. In the CRS reactor, NH₃ is introduced through a hollow needle electrode into the corona discharge zone where NH₃ molecules dissociate to NH₂ and NH radicals. These radicals react with NO_x molecules converting them into NH₄NO₃ solids. The record NO_x removal energy yield of the CRS method is 15 g/kWh at NO_x removal of 90% (Table 1, [9]). The disadvantage of the corona discharge is the fact that selective control of the plasma chemical reaction to reduce NO_x to N₂, like in the SCR, is difficult.

In recent years hybrid plasma-catalyst systems have been proposed because of their capability to reduce NO_x into N₂ [11–13,16–21]. A typical hybrid system consisted of a corona discharge and a catalyst operating at a temperature over 120 °C. So far researchers used a catalyst either as a layer coating the plane electrode in a point-to-plane dc corona discharge reactor [16], or as pellets or globules in a packed bed silent discharge reactors [17–21]. In some

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Table 1

Parameters of NO_x removal processing in selected systems using corona discharge (CD) and ammonia

System	Gas mixture	Gas temperature (°C)	Initial NO concentration (ppm)	Flow rate (l/min)	Specific energy (Wh/m ³)	NO _x removal efficiency (%)	Energy yield (g NO/kWh)	Ref.
CD (wire-to-cylinder)	N ₂ :O ₂ :CO ₂ (balance:10%:10%)	22	400	2	333	90	0.6	[5]
CD (wire-to-plate)	Flue gas from coal-burning power station	100	300	10000	12	60	20	[6]
CD (needle-to-plate)	Combustion gas from city gas burner	70–200	200	60	230	90	0.2	[7]
CRS	N ₂ :O ₂ :CO ₂ :H ₂ O:SO ₂ (89.4%:2.6%:7.9%:<0.5%:800 ppm)	22	200	5	18	90	15	[9]
Hybrid CD-catalyst	N ₂ :O ₂ :CO ₂ (85%:10%:5%)	150	430	2–6	8.3	75	21	[11]
Hybrid CD-catalyst	Diesel exhaust gas	94–189	198	1314	2.5	60	55	[13]
Hybrid CD-catalyst	N ₂ :O ₂ :CO ₂ (80%:5%:15%)	22	200	1	80	96	3.4	This work

hybrid systems, first the corona discharges were used for oxidizing NO into NO₂, and then the SCR processing with NH₃ was used to convert NO₂ into N₂ and H₂O [11–13]. The efficiency of NO_x removal using hybrid plasma-catalyst systems was up to 98% at an energy yield up to 55 g/kWh (Table 1, [13]). In other hybrid systems, hydrocarbons were added to the flue gas polluted with NO_x to improve its removal [17–19].

This work was aimed at studying NO_x removal in a hybrid system consisted of a dc streamer corona discharge with a catalyst operating under low temperature (22 °C) in the presence of NH₃. In our reactor a catalyst typical of the SCR method, but operating at low temperature, interacted with a corona discharge. Gaseous NH₃ was introduced into the corona discharge zone through a hollow needle electrode similarly as in CRS reactors. We used a new way of introducing gaseous NH₃ into the corona discharge zone. In this new method, the gaseous NH₃ was introduced into the discharge zone after releasing it by the corona discharge from an NH₃ phase, which has been earlier adsorbed on the catalyst surface. The performance of our hybrid corona discharge-catalyst reactor is described below.

2. Experimental

The hybrid corona discharge-catalyst reactor used in this experiment is shown in Fig. 1. The positive dc corona discharge was generated between a stainless steel hollow needle electrode and grounded flat mesh electrode (1 mm × 1 mm) made of brass. The hollow needle electrode was placed perpendicularly to the mesh. The outer and inner diameters of the hollow needle were 2 and 1.6 mm, respectively. The hollow needle-mesh spacing was 40 mm.

The catalyst used in this investigation was a layer of V₂O₅ and TiO₂, as in the SCR processing, deposited on Al₂O₃ 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 two layers. The hollow needle-catalyst spacing was about 30 mm.

The catalyst layer deposited on Al₂O₃ globules was used to play two roles. The first one, when a “pure” catalyst-covered globules were used, was the typical role of catalyst. The another role, consisting a novel in this investigation, resulted from using the catalyst-covered globules as an adsorber for NH₃ before placing the globules into the reactor. We expected that the NH₃-saturated globules could become a gaseous NH₃ source in the reactor, when subjected to the corona discharge. The releasing of gaseous NH₃ from the catalyst globules by the corona discharge was supposed to be an efficient way of NH₃ supply into the reactor.

In the following text, the case when the “pure” catalyst was used is called an NH₃-free catalyst case, while an NH₃-saturated catalyst case corresponds to the experiment in which the catalyst with NH₃ adsorbed on its surface was used.

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, sustaining a stable streamer corona discharge. The time-averaged discharge current varied in the range of 50–200 μA.

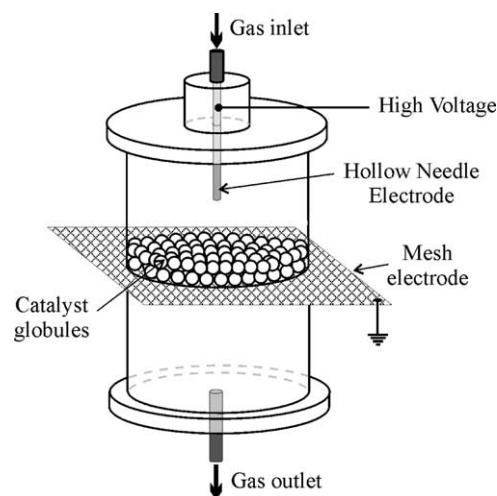


Fig. 1. The hybrid corona discharge-catalyst reactor.

A gas mixture of N₂ (80%):O₂ (5%):CO₂ (15%):NO (200 ppm), simulating a flue gas (without water and hydrocarbons), was supplied through the hollow needle and then flowed through the reactor with a flow rate of 1 l/min.

When the performance of the reactor with the “pure” catalyst was investigated, gaseous NH₃ was first mixed with the N₂:O₂:CO₂:NO mixture, forming the working gas. Then the working gas was introduced through the hollow needle electrode into the reactor. The NH₃ concentration in the working gas was 400 ppm, which corresponds to a molar ratio of NH₃ to NO equal to 2:1. As it was found by Park et al. [9], the removal efficiency of NO_x by the corona discharge is highest when the molar ratio NH₃:NO = 2:1.

When the performance of the reactor with NH₃-saturated catalyst globules was tested, gaseous NH₃ (30 ppm) was introduced into the reactor, after mixing with the N₂:O₂:CO₂:NO mixture. This additional gaseous NH₃ was supposed to make up a loss of NH₃ released from the globules and sustain steady concentration of NH₃ in the reactor with a minimum slip in the outlet gas (1 ppm). As the concentration of NH₃ adsorbed on the globules layer was expected to decrease with elapsing time of the discharge operation, this part of the investigation had a time-dependent character. Also, time-dependent deterioration of the catalyst layer when employing NH₃ was expected (a kind of catalyst poisoning by products of the plasma-chemistry reactions in the reactor).

Concentrations of NO, NO₂ and NH₃ in the working gas at the reactor inlet and outlet were measured by absorption spectroscopy method using a Perkin-Elmer 16 PC FTIR spectrophotometer operating in the infrared range 4400–1000 cm⁻¹.

3. Results

The experimental results showed that three factors influence strongly the efficiency of NO_x removal from the working gas. They are: the plasma (parametrically represented here by the energy density), catalyst and NH₃ including the way of its supplying into the reactor. The details are described below.

3.1. Corona discharge processing without catalyst and gaseous NH₃ supply

The removal efficiency of NO from the working gas by processing it with the corona discharge without catalyst and NH₃ was no higher than 25% (at energy density of 400 J/l, Fig. 2). The processing was accompanied with production of NO₂ up to 50 ppm. As a result, NO_x removal was not higher than 8% (Fig. 3). In this case, NO is believed to be removed mainly in reactions with N and O atoms and O₃ molecules, produced in the corona discharge [22]:

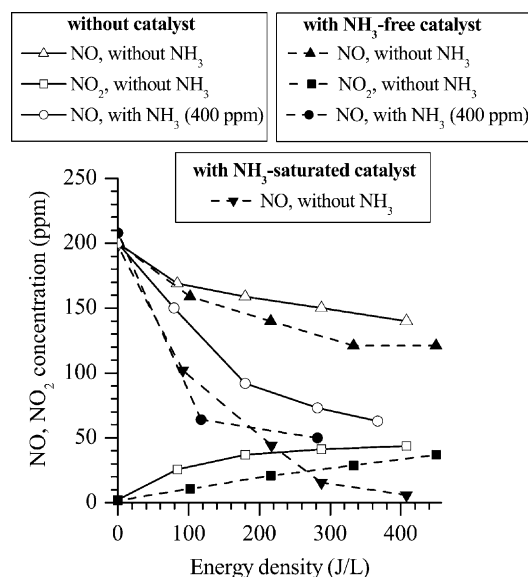


Fig. 2. Concentrations of NO and NO₂ in the working gas at the reactor outlet as a function of energy density. Cases: (a) processing without catalyst [without or with gaseous NH₃ (400 ppm) supply]; (b) processing with NH₃-free catalyst [without or with gaseous NH₃ (400 ppm) supply]; and (c) processing with NH₃-saturated catalyst [without gaseous NH₃ (30 ppm) supply].



The resulting balance between the production and decomposition of NO and NO₂ molecules is influenced by the reverse reaction:

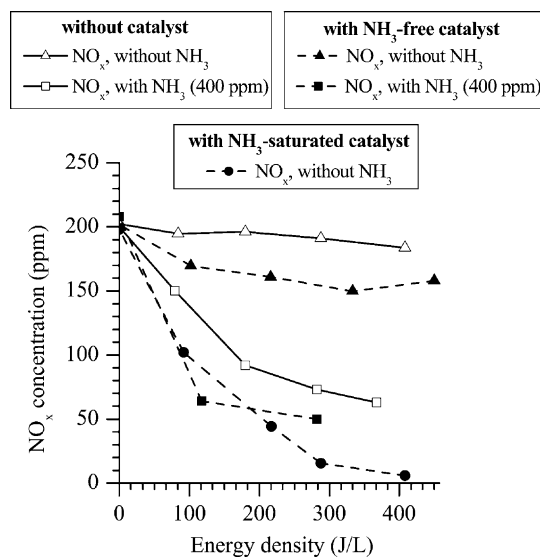


Fig. 3. Concentration of NO_x in the working gas at the reactor outlet as a function of energy density. Cases: (a) processing without catalyst [without or with gaseous NH₃ (400 ppm) supply]; (b) processing with NH₃-free catalyst [without or with gaseous NH₃ (400 ppm) supply]; and (c) processing with NH₃-saturated catalyst [without gaseous NH₃ (30 ppm) supply].

Reactions (2) and (3) are responsible for NO₂ production which causes NO_x removal low.

3.2. Corona discharge processing without catalyst and with gaseous NH₃ (400 ppm) supply

NH₃ (400 ppm) added to the working gas increased NO removal to 66% (at energy density of 400 J/l) and resulted in the absence of NO₂ molecules in the outlet gas (Fig. 2), which are consumed in reactions (5) and (6). As a result, NO_x removal (Fig. 3) is equal to NO removal.

When the working gas is processed by the corona discharge in the presence of NH₃, NO is believed to be removed in two pathways. The first pathway is oxidation to NO₂ in reactions (2) and (3). Then NO₂ reacts with OH and HO₂ radicals, produced from NH₃ and O₂ molecules in the corona discharge [9]:



Afterwards, HNO₃ reacts with NH₃ forming NH₄NO₃ solid particles:



In this experiment we could find deposits of white NH₄NO₃ solids on the reactor wall and mesh electrode produced during the corona processing. The second pathway is reduction of NO by NH₂ and NH radicals, produced in the corona discharge from NH₃ molecules [9]:



At this moment we are not able to determine the relative importance of both pathways.

Although, NH₃ was involved in both pathways of NO removal, only a small fraction, i.e. about 15%, of the introduced NH₃ is consumed during the corona discharge processing (Fig. 4). This resulted in a high slip of NH₃. It seems that the initial concentration of NH₃ used in this experiment after [9] was too high. However, we did not investigate this problem further.

3.3. Corona discharge processing with NH₃-free catalyst and without gaseous NH₃ supply

In the presence of the NH₃-free catalyst in the reactor, NO removal from the working gas without NH₃ was higher (about 40% at 340–450 J/l) than that without catalyst and NH₃ (Fig. 2). The NO removal was accompanied with NO₂ production, which is, however, lower than that without catalyst and NH₃. As a result, NO_x removal increased up to 25% (Fig. 3).

The presence of NO₂ in the post-processing gas proves that NO₂ was produced in reactions (2) and (3), causing NO

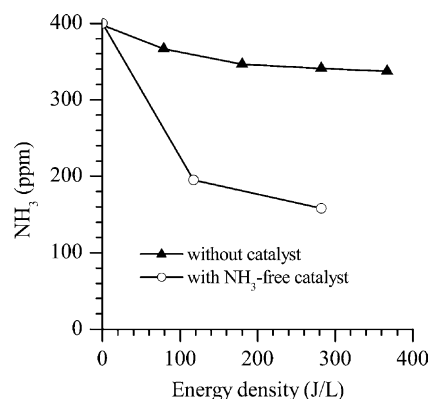


Fig. 4. Concentration of NH₃ in the working gas at the reactor outlet as a function of energy density. Inlet NH₃ concentration 400 ppm. Cases: (a) processing without catalyst [with gaseous NH₃ (400 ppm) supply]; and (b) processing with NH₃-free catalyst [with gaseous NH₃ (400 ppm) supply].

removal. However, the resulting NO₂ concentration, which was lower than that without catalyst, may result from:

- lower operating voltage (32.5 kV against 34 kV due to the shorter discharge gap with the catalyst at the same discharge current equal to 200 μA); it may cause lower production of radicals involved in NO₂ production [reactions (2) and (3)];
- losses of NO₂ molecules due to their adsorption on the catalyst surface, resulting in the lower NO₂ concentration in the outlet gas.

The higher efficiency of NO removal in the presence of the catalyst seems to be caused by the lower concentration of NO₂ molecules, which easily reduce to NO.

3.4. Corona discharge processing with NH₃-free catalyst and gaseous NH₃ (400 ppm) supply

When 400 ppm of NH₃ was introduced into the reactor with NH₃-free catalyst globules, removal of NO and NO_x increased up to 75% at energy density of 450 J/l (Figs. 2 and 3). Like in the case without catalyst, NH₃ in the working gas caused absence of NO₂ in the outlet gas due to reactions (5) and (6). Consumption of NH₃ in the processing increased with increasing energy density and reached 60% at 290 J/l (Fig. 4).

We believe that the dominant mechanisms of NO, NO₂ and NH₃ removal remain those described by reactions (5)–(9). However, the observed increase of removal efficiency of NO, NO₂ and NH₃ may be attributed to the cumulative effect of plasma enhanced reactions in gas volume, adsorption of certain gaseous compounds on the catalyst surface and reactions with these adsorbed compounds.

The importance of reactions (5) and (6) in the plasma-chemistry kinetics in the reactor with NH₃-free catalyst in the presence of NH₃ was confirmed by production of NH₄NO₃ particles [according to reaction (7)], which deposited in the form of solids on the catalyst globules and

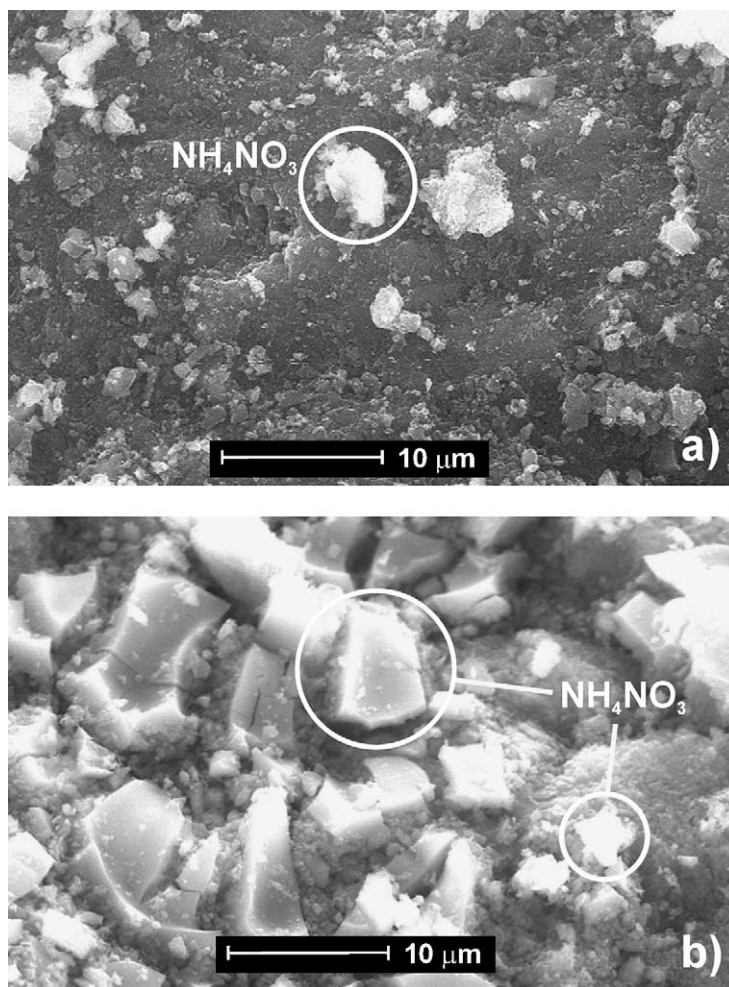


Fig. 5. SEM images of the catalyst surface, (a) after short-time (5 min) processing with NH_3 -free catalyst and gaseous NH_3 (400 ppm) supply; and (b) after long-time processing (67 h) with NH_3 -saturated catalyst and NH_3 (30 ppm) supply. Magnification: 2000 \times . Irregular and crystal forms of NH_4NO_3 are clearly present on the catalyst surface.

the reactor wall. An image of the catalyst surface taken using scanning electron microscopy (SEM) after short-time corona processing (5 min) is presented in Fig. 5a. As it can be seen, the number of NH_4NO_3 solid deposits on the catalyst surface was relatively low. The NH_4NO_3 solids had the form of irregular conglomerates (like incipient crystals), the size of which is approximately 5 μm . However, after longer time of the processing the catalyst surface became considerably deteriorated with NH_4NO_3 deposits, which resulted in a progressive decrease of NO removal. Eventually, the deteriorated catalyst surface stopped acting as a catalyst.

3.5. Corona discharge processing with NH_3 -saturated catalyst without gaseous NH_3 supply

When the catalyst saturated with NH_3 without any additional supply of gaseous NH_3 was used, removal of NO and NO_x increased to 96% (Figs. 2 and 3). NO_2 molecules were not present in the outlet gas, which, however, contained 2 ppm of NH_3 . This NH_3 slip was formed by those

NH_3 molecules, which released from the catalyst surface and were not consumed in reactions (7)–(9).

The very high efficiency of NO (and NO_x) removal recorded in this case suggests that the surface reactions with adsorbed compounds are very efficient when the interaction of the corona discharge with the NH_3 -saturated catalyst takes place.

The high NO (and NO_x) removal and relatively low discharge power resulted in a high energy yield of the NO removal process. At NO removal efficiency of 96% and discharge power of 4.8 W the energy yield was 3.4 g NO/kWh (Table 2). This is superior to the other cases tested in this experiment (Table 1). However, better results were obtained in other hybrid plasma-catalyst [13] and CRS systems [9].

The number and shape of NH_4NO_3 solids deposited on the catalyst surface are similar to those in Fig. 5a, which corresponds to the case of NH_3 -free catalyst and gaseous NH_3 supply.

After some time, the NH_3 -saturated catalyst started to lose its properties due to either NH_3 depletion on the

Table 2

Parameters of NO_x removal processing in the hybrid corona discharge-catalyst reactor

Processing	Discharge voltage (kV)	Discharge power (W)	Removal efficiency (%)	Energy yield (g NO/kWh)
Without catalyst and gaseous NH ₃ supply	34	6.8	25	0.7
Without catalyst, with gaseous NH ₃ (400 ppm) supply	31	6.2	66	1.8
With NH ₃ -free catalyst, without gaseous NH ₃ supply	32.5	6.5	40	1.0
With NH ₃ -free catalyst and gaseous NH ₃ (400 ppm) supply	29	5.8	75	2.2
With NH ₃ -saturated catalyst, without gaseous NH ₃ supply	24	4.8	96	3.4

Flow rate: 1 l/min; corona discharge current: 200 μ A.

catalyst surface or deterioration of the catalyst surface by NH₄NO₃ solids. This was investigated in the following experiment.

3.6. Temporal performance of the hybrid reactor

Fig. 6 illustrates temporal performance of the hybrid reactor for the following cases:

- processing without catalyst [without or with gaseous NH₃ (400 ppm) supply];
- processing with NH₃-free catalyst [without gaseous NH₃ (400 ppm) supply];
- processing with NH₃-saturated catalyst [without or with gaseous NH₃ (30 ppm) supply].

In all the cases the discharge current was fixed at 100 μ A.

As expected, NO concentration at the reactor outlet remained constant when the working gas processing was carried out either without catalyst or with NH₃-free catalyst without gaseous NH₃ supply.

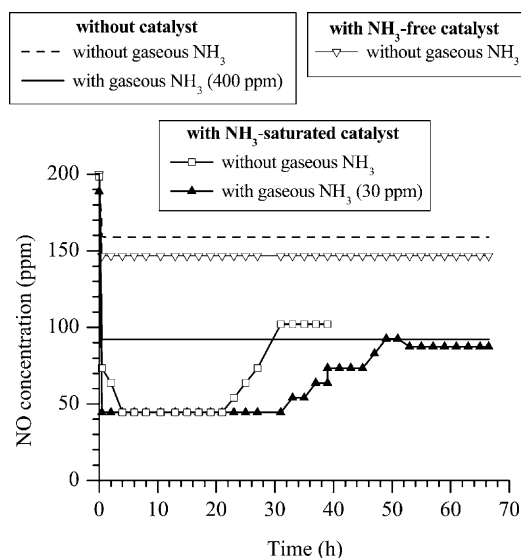


Fig. 6. Time dependence of NO concentration in the working gas at the reactor outlet. Corona discharge current 100 μ A. Cases: (a) processing without catalyst [without or with gaseous NH₃ (400 ppm) supply]; (b) processing with NH₃-free catalyst [without gaseous NH₃ (400 ppm) supply]; and (c) processing with NH₃-saturated catalyst [without or with gaseous NH₃ (30 ppm) supply].

When the NH₃-saturated catalyst without gaseous NH₃ supply was used, NO (and NO_x) removal efficiency was relatively high (78% at a discharge current of 100 μ A) and remained constant over the first 20 h. After that time, the NH₃-saturated catalyst started to loose its activity and NO removal efficiency decreased. After the next 10 h, the NO removal efficiency remained steady at a level corresponding to the corona discharge processing without catalyst but with gaseous NH₃ (400 ppm) supply.

When the corona discharge processing was performed in the presence of NH₃-saturated catalyst with gaseous NH₃ (30 ppm) supply, NO removal efficiency was equal to that without gaseous NH₃ supply (78%) but it remained constant longer (during about 30 h). After that time the catalyst lost its activity. As a result, NO removal efficiency started to decrease and NO concentration increased during the next 20 h until it leveled at a concentration typical of the processing without catalyst but with gaseous NH₃ (400 ppm) supply. After leveling at a time of 50 h, the performance of the hybrid reactor became unchanged over the next 20 h.

Without additional gaseous NH₃ supply catalyst lost its activity 10 h earlier then in the case with gaseous NH₃ (30 ppm) supply (Fig. 6). It proves that during corona discharge processing NH₃ molecules are released from the NH₃-saturated catalyst surface.

The significant changes of the catalyst surface due to NH₄NO₃ deposits after 67 h corona discharge processing were revealed by scanning electron microscopy (SEM) imaging (Fig. 5b). The catalyst surface exposed to the corona discharge was almost completely covered with NH₄NO₃ solids, which formed relatively large crystals.

Taking the above into consideration, we conclude that the NH₃-saturated catalyst (without or with gaseous NH₃ supply) lost its activity not due to releasing NH₃ from its surface but due to NH₄NO₃ deposits. The surface-deteriorated catalyst still was able to release NH₃ molecules enough to remove NO molecules on a level typical of processing with gaseous NH₃ (400 ppm) supply.

From the weight balance analysis of the NH₃ and NH₄NO₃ carried out after long-time processing [NH₃-saturated catalyst with gaseous NH₃ (30 ppm) supply], we found that only a small fraction of NO and NO₂ was transformed into NH₄NO₃, according to reaction (7). This resulted from a small quantity of NH₄NO₃ solids weighted after the processing, compared to that predicted

by Eqs. (7)–(9), if they were the only pathway of NO and NO₂ removal. This may mean that part of NO and NO₂ are reduced into molecular nitrogen via other reactions related to the presence of ammonia.

4. Summary and conclusions

In this paper results of investigation of the performance of a hybrid corona discharge-catalyst reactor to be used for NO_x removal from the flue gases are presented. The catalyst was a layer of V₂O₅ and TiO₂ deposited on Al₂O₃ globules. The gas mixture, which simulated a flue gas, was N₂ (80%):O₂ (5%):CO₂ (15%):NO (200 ppm). The corona discharge processing of the gas mixture was carried out: without catalyst, with NH₃-free catalyst, and with NH₃-saturated catalyst, all either without or with gaseous NH₃ supply.

The investigation showed that:

- The catalyst increased significantly NO_x removal efficiency. The highest NO_x removal (96% at energy density of 400 J/l) was observed when the catalyst was saturated with NH₃ [case without gaseous NH₃ (30 ppm) supply].
- In the presence of NH₃, the corona discharge produced solid particles of NH₄NO₃ which depositing on the catalyst surface shortened the active life of the catalyst (poisoning of the catalyst). The life-time of the catalyst due to its poisoning was about 30 h.
- NO_x removal energy yield in the presented reactor with NH₃-saturated catalyst without additional gaseous NH₃ supply is up to 3.4 g/kWh (at 96% of NO_x removal) compared to 1.8 g/kWh in the “pure” corona discharge processing with gaseous NH₃ (400 ppm) supply.
- The hybrid reactor with NH₃-saturated catalyst operated efficiently over 30 h without additional gaseous NH₃ supply.

The obtained results showed that the proposed hybrid reactor consisted of the corona discharge and NH₃-saturated catalyst is attractive for NO_x removal and may be alternative to other non-thermal plasma methods used for NO_x removal. The processes which occur when the corona discharge interacts with NH₃-saturated catalyst are not known and need

further investigation the results of which may improve performance of the presented reactor.

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