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Experimental study of the NO and NO_2 degradation by photocatalytically active concrete

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ABSTRACT

The application of photocatalytic concrete containing TiO_2 in urban streets is a method to improve the air quality in highly polluted areas. By using this technology it is possible to degrade a wide range of air contaminants, like nitric oxide (NO) and nitrogen dioxide (NO_2), mainly emitted by automobiles. In the present paper, the photocatalytic degradation of NO and NO_2 is experimentally studied, and the atmospheric reactions involving nitrogen oxides and solar radiation are analyzed as well. In addition, the influence of different system parameters, such as inlet pollutant concentration, relative humidity, and irradiance is investigated in detail.

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1. Introduction

Heterogeneous photocatalytic oxidation is a promising and sustainable technology to be applied to water and air purification. This process applies a solid semiconductor catalyst, normally titanium dioxide (TiO_2), which is activated by ultraviolet (UV) light of certain wavelength. The main advantages of this technology are: (i) pollutants can be completely transformed into innocuous products, and (ii) a wide range of contaminants can be treated due to its very low selectivity.

Nowadays, nitrogen oxides (NO_x) represent a serious environmental problem in highly contaminated areas. Nitrogen oxides (mainly NO and NO_2) are produced during fuel burning processes, for example in automobiles, causing a wide variety of health and environmental impacts, like the formation of tropospheric ozone and urban smog through photochemical reactions with hydrocarbons. Furthermore, NO_x together with SO_x (sulfur dioxide and sulfur trioxide) are the major contributors to the "acid rain".

One of the European Union (EU) directives (1999/30/EC) [1] established limit values for concentrations of the most representative air pollutants. However, NO_x exceeds the maximum allowed limit, especially in urban areas (e.g. in the Randstad conurbation, the Netherlands [2]), and extra measures are needed to solve this problem. The application of TiO_2 in concrete pavement has shown a promising effect in the removal of nitrogen oxides and attempts to improve the air quality in urban streets [3–5].

Currently, there are some international and national standards available to assess the photocatalytic oxidation efficiency of semiconducting materials. Some of them employ only NO as the target pollutant [6], while others use a mixture of NO and NO_2 to perform the test [7,8]. However, since the test is carried out in the presence of light, not only photocatalytic reactions take place but also homogeneous and photolysis reactions of NO_x occur, but so far, no research has linked this to their photocatalytic oxidation.

In the present paper, the photocatalytic degradation of NO and NO_2 is experimentally studied and the homogeneous reactions involving NO_x are analyzed employing a photocatalytic concrete stone, and a normal non-active concrete block as a blank sample. In addition, the influence of different system parameters, such as inlet pollutant concentration, relative humidity, and irradiance is investigated. Depending on the employed operating conditions, part of the NO_x conversion employing the photocatalytic stone corresponds to homogeneous reactions under UV light [9], but not just to photocatalytic reactions alone.

2. Experimental

The applied experimental set-up is composed of a continuous planar reactor housing the concrete stone sample, a suitable UV-A light source, a chemiluminescent NO_x analyzer, and an appropriate gas supply system for NO and NO_2 (Fig. 1). Table 1 shows the main characteristics, dimensions and operating conditions of the test setup employed to execute the NO_x degradation experiments.

To carry out the experiments, one photocatalytic and one normal commercial paving stone samples were used. The employed photocatalytic sample is a double layer stone with the upper layer being photocatalytically active. This upper layer is prepared by mixing

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Nomenclature

A	Area (dm^2)
B	Reactor width (dm)
C	Concentration (ppm)
E	Radiative flux ($W\,m^{-2}$)
H	Reactor height (dm)
L	Reactor length (dm)
Q	Flow rate ($l\,min^{-1}$)
RH	Relative humidity (%)
t	Time (min)
V	Reactor volume (dm^3)

Subscripts

in	Inlet condition
NO	Nitric oxide
NO_2	Nitrogen dioxide
NO_x	Nitrogen oxides
out	Outlet condition

Table 1
Experimental setup main characteristics and operating conditions.

Description	Operating conditions
Reactor	
Length (L)	2 dm
Width (B)	1 dm
Height (H)	0.03 dm
Volume (V)	0.06 dm^3
Photocatalytic/blank concrete stone	
Length (L)	2 dm
Width (B)	1 dm
UV Lamps: Philips Compact S × 3	
Input power	25 W
Emission wavelength	300–400 nm
Flow rate (Q)	3 $l\,min^{-1}$
Relative humidity (RH)	10–70%
Irradiance flux (E)	2–11 $W\,m^{-2}$
NO inlet concentration ($C_{NO,in}$)	0–1 ppm
NO_2 inlet concentration ($C_{NO_2,in}$)	0–1 ppm

dry powders of normal concrete with TiO_2 powder before adding water. According to X-ray fluorescence (XRF) analysis performed in the upper layer of the stone, the present concrete sample has a TiO_2 content of 5.9% by weight. In preparation of each experiment, the sample surface is washed with water in order to remove fouling, contamination and potential reaction products due to a previous NO_x degradation.

After assembling the sample the reactor is closed and the gas supply is started. The UV-A source is switched on as well in order to stabilize the radiation, but the reactor stays covered to prevent the first degradation. With the help of the controls, the flow rate and the relative humidity are adjusted. The supplied NO/NO_2 concentration is adjusted to the desired inlet concentration, which is monitored by the analyzer. When these conditions are stable the data acquisition is started. After this period of time, the reactor cover is removed to allow the UV-radiation passing through the glass. The degradation for the uncovered reactor lasts for 20 min, then the reactor is covered again and the experiment is continued for another 5 min. Within the last minutes of measurement the NO and NO_x concentrations return ideally to the original scale. An example of a representative experimental result is shown in Fig. 2, where the different steps mentioned above and the degradation of NO and NO_2 can be observed.

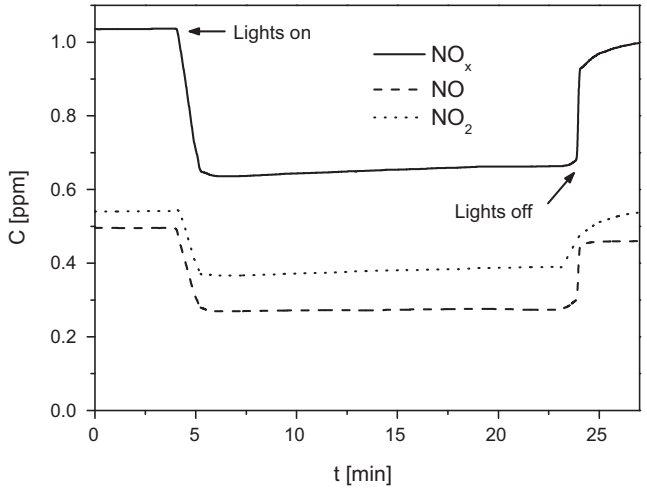

Fig. 2. Representative experimental result. $H = 3$ mm, $Q = 3\,l\,min^{-1}$, $C_{NO_2,in} \sim 0.5$ ppm, $C_{NO,in} \sim 0.5$ ppm, $E = 10\,W\,m^{-2}$ and RH = 50%.

3. Results

Table 2 shows the results of NO/NO_2 conversion and the operating conditions employed in every performed experiment. In this table, four types of experiments are listed: (i) experiments feeding only NO to the system; (ii) experiments employing only NO_2; (iii) experiments employing a mixture of NO and NO_2, and (iv) blank experiments using a non-photocatalytic sample. Within these groups, different experiments were carried out by varying the inlet concentration of NO or NO_2 and operating conditions of irradiance and relative humidity.

Figs. 3–7 show the obtained results under different operating conditions. Figs. 3–5 show the experimental results of NO and NO_2 outlet concentration with varying the inlet concentration of the pollutants, the relative humidity and the irradiance when NO, NO_2

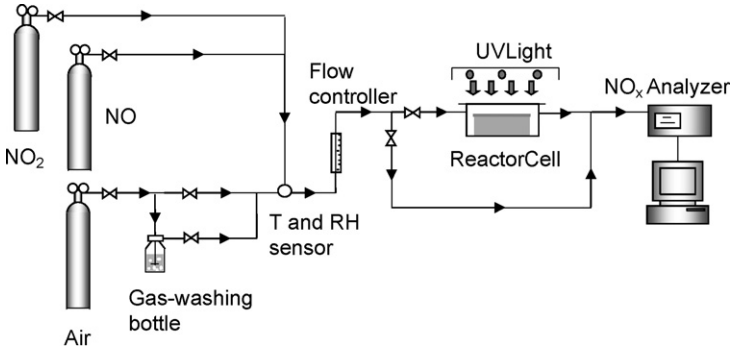

Fig. 1. Schematic representation of the experimental setup.

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