Integrated NO_x and particulate matter photocatalytic removal process

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Background

Emissions of diesel engines contain particulate matter (PM) and nitrogen oxides (NO_x) which have a negative influence on human health and the environment. Consequently an integrated system to remove both pollutants is important.

A wide range of exhaust treatment technologies has been developed e.g. diesel particulate filters (DPF) and continuously regenerating trap (CRT) catalysts which oxidize NO to NO_2, a stronger oxidizing agent for the PM degradation [1,2]. The regeneration of the DPF systems can be problematic, while the CRT systems have a poor abatement of NO_x. Systems for combined removal of both pollutants, like perovskite-type oxides, are still limited to lab-scale research [3,4].

Photocatalysis could be the solution to overcome these problems. Illumination activates the photocatalytic material (e.g. TiO_2) if $h\nu \geq$ the bandgap (E_g) of the material. Due to generation of electron-hole pairs, oxidation and reduction of organic pollutants and NO_x is accomplished. The photocatalytic removal process is based on the same principal as CRT, using NO_2 as oxidizing agent for degradation of PM, but could have the additional advantage of reducing NO_x to N_2 in order to solve the NO_x problem [5,6].

The main objective of this research is to study the reaction mechanism occurring on photocatalytic titanium dioxide materials during the integrated removal of NO_x and PM.

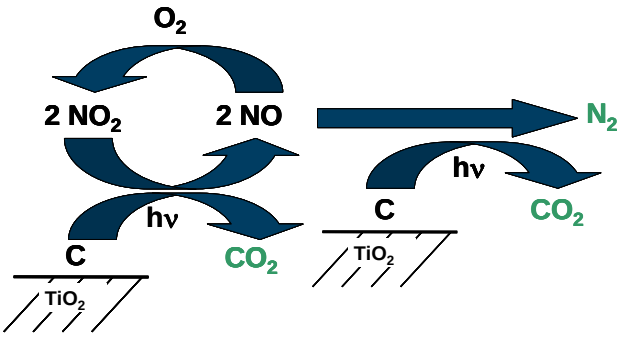

Figure 1: Proposed reaction mechanism for the photocatalytic removal of NO_x and PM

Investigation methods and results

At the laboratory for Sustainable Energy and Air Purification (University of Antwerp), in-house developed titanium dioxide based photocatalysts are effective for the conversion of NO. In Figure 2, a concentration of 250 ppm NO in a flow of 400 cm^3/min (20% O_2 and 10% H_2O) was successfully removed when UV-light activated the material. In addition to the reduction of NO_x, photocatalysis with titanium dioxide has already proven to be successful for the abatement of various contaminants and it is active at room temperature. Ceramic photocatalytic foams which can trap PM and allow simultaneously the catalytic combustion of PM and the degradation of NO_x are being studied. Preliminary tests in our laboratory confirm the possibility to remove model soot (Printex-U, Evonik) deposited on titanium dioxide pellets illuminated with UV-C light (254 nm). With Fourier transform infrared (FTIR) spectroscopy and gas sensors, the removal reactions can be analyzed and possible intermediates can be monitored.

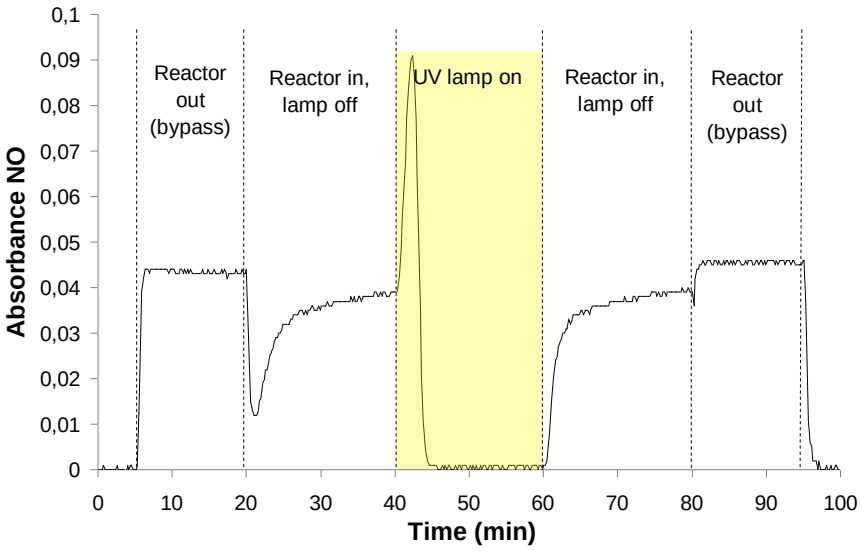

Figure 2: Photocatalytic conversion of 250 ppm NO in a flow of 400 cm³/min (20% O_2 and 10% H_2O)

Conclusions

The first results confirm the possibility to remove NO_x and PM with photocatalysis. Further research will focus on the removal efficiencies of a combined photocatalytic removal process. A diesel generator set-up is in development to confirm these results with real exhaust gases.

A deeper insight in the ongoing reactions is important to achieve a real breakthrough in photocatalytic removal processes. These processes will be analyzed under a wide range of varying conditions and the obtained data will be used to reveal the reaction mechanism. This will lead to fundamental knowledge that can help to improve materials and optimize reaction conditions. A breakthrough in this research area would have a major impact on a social, economical and technical level.

References

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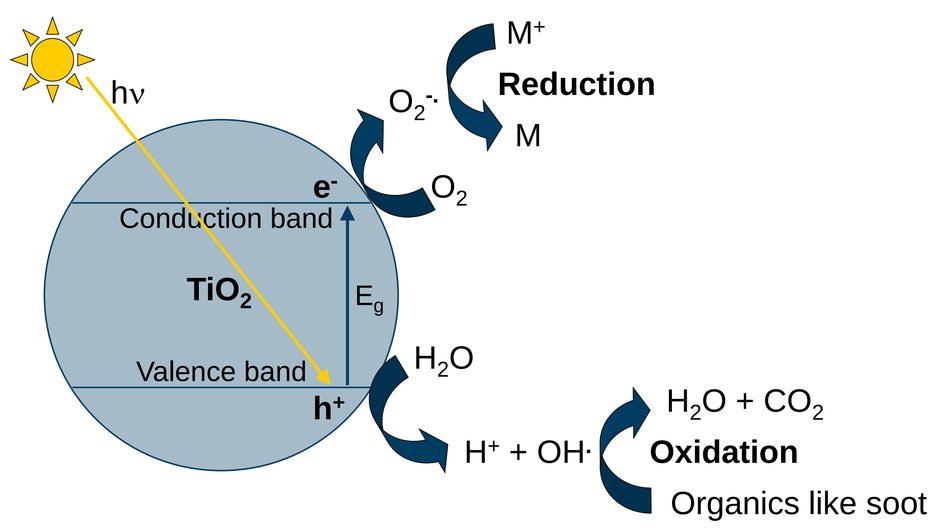
Introduction

Emissions of diesel engines contain **particulate matter (PM)** and **nitrogen oxides (NO_x)** which have a negative influence on human health and the environment. Existing exhaust treatment technologies have their disadvantages. Continuously regenerating trap (CRT), for instance, oxidizes NO to NO_2, which is a strong oxidizing agent for the PM degradation. But CRT has a poor NO_x abatement. Other systems for combined removal of both pollutants, like perovskite-type oxides, are still limited to lab-scale research.

➔ An **integrated photocatalytic removal process** will be studied to solve these problems.

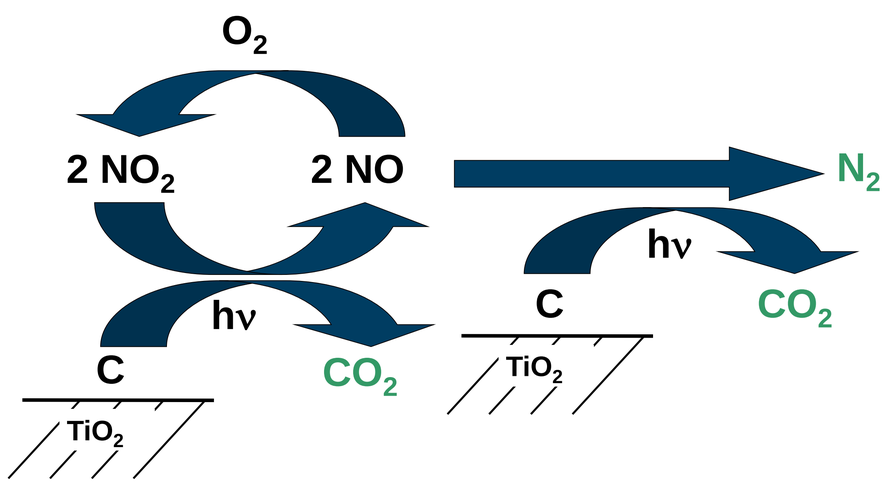
Photocatalysis

Illumination activates the photocatalytic material (e.g. TiO_2) if $h\nu \geq$ the bandgap (E_g) of the material. Due to the activation, oxidation and reduction of organic pollutants is accomplished.



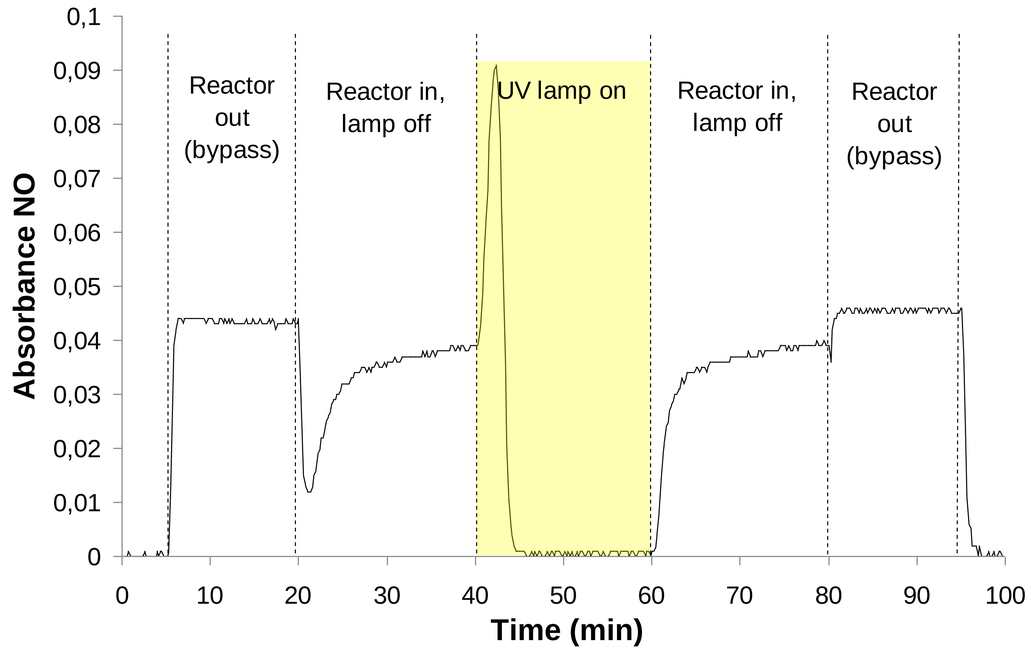
Reaction mechanisms

Like CRT, a photocatalytic removal system uses **NO_2 as oxidizing agent** for the degradation of PM.
Photocatalysis could have the additional advantage of reducing NO_x to N_2 which would solve the NO_x problem [1,2].



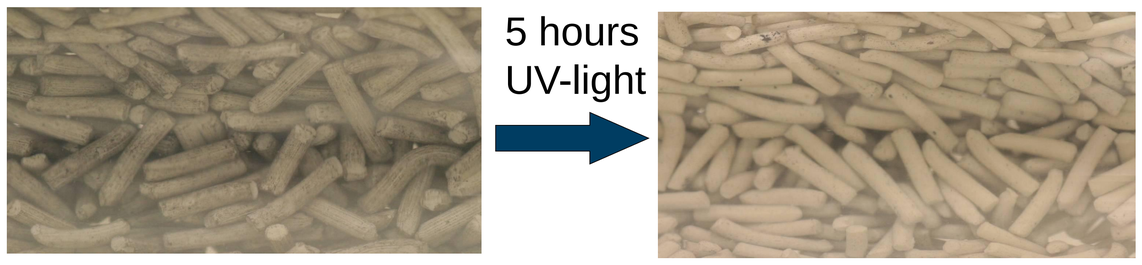
Result: The degradation of NO_x

In-house developed reactors with photocatalytic material are effective for the conversion of NO. In the graph, a concentration of 250 ppm **NO** in a flow of 400 cm^3/min (20% 0_2 and 10% H_2O) was **successfully removed** when UV-light activated the material.



Result: The degradation of PM

Preliminary tests in our laboratory confirm the possibility to remove a **model soot** (Printex-U, Evonik) deposited on titanium dioxide pellets illuminated with UV-C.



Prospectives

➔ Determination of the removal efficiencies for the combination of NO_x and PM.
➔ A set-up with a diesel generator is in development to confirm these results with real exhaust gases.
➔ Optimization of photocatalytic material and illumination.
➔ Gaining insight in the reaction mechanisms.

Take home message: YES to photocatalysis

Yes, photocatalysis can degrade NO_x and PM successfully.

Further research on the reaction mechanisms and performance of the system is necessary.

References [1] Poulston et al., Appl. Cat. B (2009), 89, 335-341 [2] Zhang et al., Journ. Cat. (2001), 198, 1-8

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