

PHOTOCATALYTIC APPLICATIONS IN BELGIUM, PURIFYING THE AIR THROUGH THE PAVEMENT

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ABSTRACT

The use of materials can influence to a large extent the environmental impact of traffic and of road infrastructure. Especially at urban areas, where the risk of smog formation during hot summer days is high, the use of photocatalytic pavements can reduce the air pollution significantly.

A first application on the side roads of the main entrance axis in Antwerp has been studied by the Belgian Road Research Centre [1]. Although measurements on site did not give the expected results due to the configuration of the road surface, the laboratory results indicated a good efficiency towards NO and NO₂ abatement as well as a good durability of this air cleaning characteristic.

New applications in industrial zones (in the framework of an INTERREG project) as well as in the tunnels of Brussels (LIFE⁺ project) are put in place in order to measure the influence on site.

The photocatalytic material, titanium dioxide, is introduced in the top layer of the concrete pavement. In contact with light, TiO₂ is activated and able to reduce different pollutants in the air. Special attention is given to the NO and NO₂ content in the air, since they are for over 50% caused by the exhaust of traffic and are at the base of smog, secondary ozone and acid rain formation.

This paper first describes the principle of photocatalytic materials through the results of the research project(s). Subsequently, the different applications in Antwerp (2005) and at the industrial zone in Wijnegem (2011) will be described. In the latter, a double layered concrete is used. By using this configuration, the TiO₂ can be added to the top layer, while the bottom layer is made of recycled concrete aggregates in order to reduce the ecological impact.

Keywords: TiO₂; photocatalysis; air purification; concrete pavement; surface treatment

1. INTRODUCTION

Emission from the transport sector has a particular impact on the overall air quality because of their rapid rate of growth: goods transport by road in Europe has increased by 54% since 1980, while in the past 10 years passenger transport by road in the EU has gone up by 46% and passenger transport by air by 67%. The main emissions caused by motor traffic are nitrogen oxides (NO_x), hydrocarbons (HC) and carbon monoxide (CO), accounting for respectively 58%, 50% and 75% of all such emissions [2].

These pollutants have an increasing impact on the urban air quality. In addition, photochemical reactions resulting from the action of sunlight on NO₂ and VOC's (volatile organic compounds) lead to the formation of 'photochemical smog' and ozone, a

secondary long-range pollutant, which impacts in rural areas often far from the original emission site. Acid rain is another long-range pollutant influenced by vehicle NO_x emissions and resulting from the transport of NO_x , oxidation in the air into NO_3^- and finally precipitation of nitric acid with harmful consequences for building materials (corrosion of the surface) and vegetation.

The European directives impose a limit to the NO_2 concentration of max. $40 \mu\text{g}/\text{m}^3$ NO_2 (33 ppbV) averaged over 1 year and $200 \mu\text{g}/\text{m}^3$ (163 ppbV) averaged over 1 hour. These limit values gradually decreased from 50 and 250 in 2005 to the final limit in 2010 [3].

Heterogeneous photocatalysis is a promising method for NO_x abatement. As will be indicated in the last paragraph of this paper, different applications in Belgium are also starting to be implemented. Up till now, UV-light was necessary to activate the photocatalyst. However, recent research indicates a shift towards the visible light. This means that applications in tunnels and indoor environments become more realistic. Especially the application in tunnels is worth looking at due to the concentration of air pollutants at these sites. One of the projects in Belgium is focusing on this subject [4].

2. HETEROGENEOUS PHOTOCATALYSIS, A PROCESS FOR AIR PURIFICATION

A solution for the air pollution by traffic can be found in the treatment of the pollutants as close to the source as possible. Therefore, photocatalytic active materials can be added to the surface of pavement and building materials. In combination with (sun)light, the pollutants are oxidized, due to the presence of the photocatalyst and precipitated on the surface of the material. Afterwards, they can be removed from the surface by the rain or cleaning/washing with water (see Figure 1).

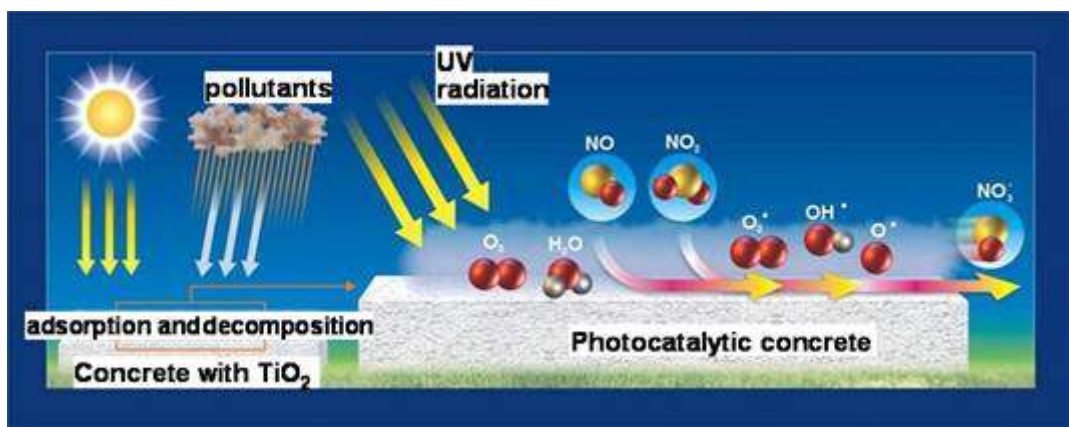


Figure 1 – Schematic of photocatalytic air purifying pavement (CTG).

Heterogeneous photocatalysis with TiO_2 as catalyst is a rapidly developing field in environmental engineering. It has a great potential to cope with the increasing pollution. The impulse for the use of TiO_2 as photocatalyst was given by Fujishima and Honda in 1972 [5]. They discovered the hydrolysis of water in oxygen and hydrogen in the presence of light, by means of a TiO_2 -anode in a photochemical cell. In the eighties, organic pollution in water was decomposed by adding TiO_2 and under influence of UV-light (with wave lengths lower than 387nm). The application of TiO_2 , in the photoactive crystal form anatase, as air purifying material originated in Japan in 1996. Since then, a broad spectrum of products appeared on the market for indoor use as well as for outdoor use. In the case of traffic environments, it is important that the exhaust gasses stay in contact with

the surface during a certain period. The geometrical situation, the speed of the traffic, the speed and direction of the wind, the temperature, all influence the final reduction rate of pollutants in situ.

In the case of concrete pavement blocks [6-7], the anatase is added to the wearing layer of the paver which is approximately 8 mm thick. The fact that the TiO_2 is present over the whole thickness of this layer means that even if some abrasion takes place by the traffic, new TiO_2 will be present at the surface to maintain the photocatalytic activity. Another, similar application consists of using a double layered concrete with addition of TiO_2 (in the mass and/or as dispersion on the surface) to the top layer which will be discussed later on. The use of the TiO_2 in combination with cement leads to a transformation of the NO_x into NO_3^- , which is adsorbed at the surface due to the alkalinity of the concrete, thereby creating a synergetic effect. Subsequently, the deposited nitrate will be washed away by rain.

3. LABORATORY RESULTS: PARAMETER EVALUATION

Different test methods have been (and still are being) developed to determine the efficiency of photocatalytic materials towards air purification. An overview is given in [8]. A distinction can be made by the type of air flow. In the flow-through method according to ISO 22197-1 (2007) [9], the air, with a concentration of 1 ppmV of NO , passes over the sample, which is illuminated by a lamp with light intensity equal to 10 W/m^2 in the range between 300 and 400 nm, as illustrated in Figure 2. Afterwards, the NO_x concentration is measured at the outlet. It is also worth to note here that within Europe actions are underway to harmonize and develop new standards for photocatalysis [10]. Specifically concerning NO_x abatement, investigations are currently being made into a new type of mixed reactor system which could offer some advantages in the future. In any case, the test procedure used in this research is following the existing ISO-standard.

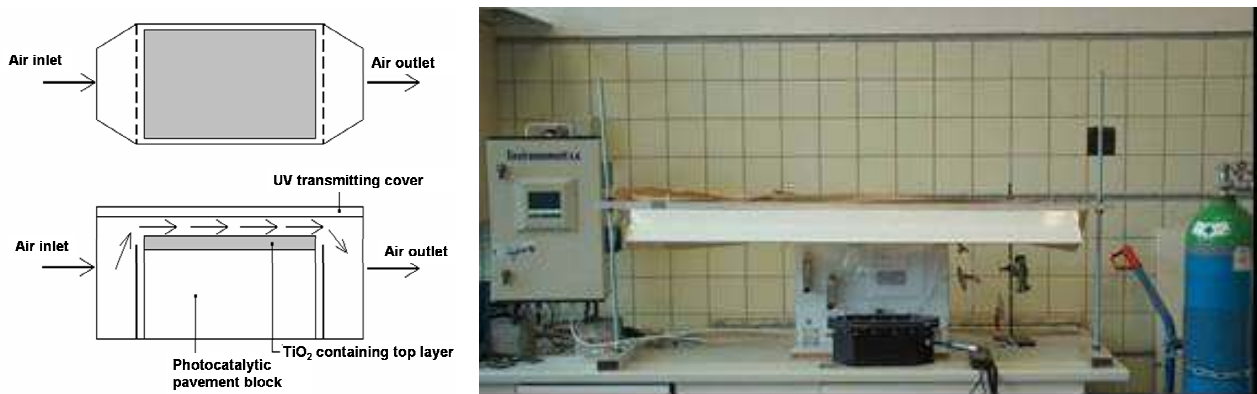


Figure 2 – Schematic and photo of measurement set-up according to ISO 22197-1 (2007) at the BRRC.

The preparation of the samples is of great importance. Due to the photocatalytic activity, NO_3^- is deposited on the surface of the material and covers to a certain extent the TiO_2 from the light and from the pollutants. By this the efficiency is lowered over time, but by rinsing the surface, the initial efficiency can be restored again, as also demonstrated later on (see part 4). The pre-treatment of the samples in the laboratory can be important to obtain reproducible results. This depends on the type of base material. In the case of concrete, the release of NO and NO_x prior to the photocatalytic reaction is limited, in the case of paints, this can be more important. The schedule of the pre-treatment is

represented in Figure 3, where a typical test scheme according to the ISO standard, is applied to the sample: 0.5 hour at 1ppmV NO-concentration, no light – 5 h exposure to an air flow of 3l/min with 1 ppmV NO-concentration and UV-illumination – 0.5 hour with UV-illumination and no exposure. A small increase in time of the NO_x concentration is visible due to the deposit of the NO₃⁻ at the surface.

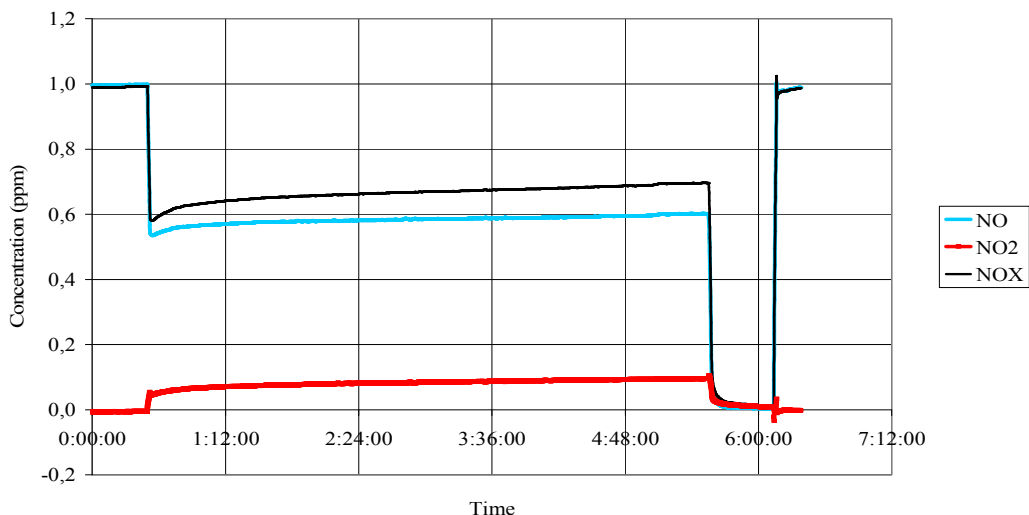


Figure 3 - Results obtained in laboratory according to the standard test procedure.

In the laboratory, the influence of different important test parameters affecting the photocatalytic reaction has been investigated such as temperature, light intensity, relative humidity, contact time (surface, flow velocity, height of the air flow over the sample...). For instance, the effect of relative humidity of the ingoing air is illustrated in Figure 4. Clearly, the reduction of NO_x concentration in the outlet air decreases with increasing relative humidity (RH). This has to do with the fact that the water in the atmosphere plays a role in the adhesion of the pollutants at the surface and the competition effect that can arise between water molecules and NO_x in the air with increasing RH. Hence, relative humidity of the air is an important limiting factor for photocatalytic applications in humid areas like Belgium.

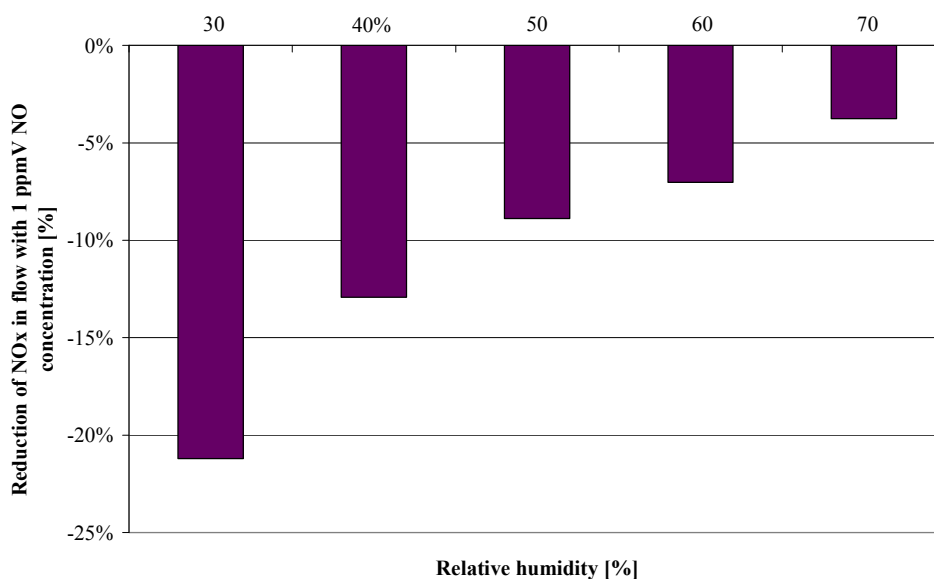


Figure 4 – Effect of relative humidity on photocatalytic efficiency

In general, it can be stated that the efficiency towards the reduction of NO_x increases with a longer contact time (larger surface area, lower air velocity, smaller height of air flow, higher turbulence at the surface), a lower relative humidity and a higher intensity of incident light. These are the conditions at which the risk of ozone formation is the largest: high temperatures, no wind and no rain. At these days, the photocatalytic reaction will be more pronounced.

4. PILOT PROJECT IN ANTWERP

An important issue is the conversion of the results, obtained in the laboratory to real applications. In order to see the influence of photocatalytic pavements in real conditions, a first pilot section of 10.000 m² of photocatalytic pavement blocks was constructed in 2004-2005 on the parking lanes of a main axe in Antwerp. Figure 5 depicts a view of the parking lane, where the photocatalytic concrete pavement blocks have been applied. Only the wearing layer of the blocks contains TiO₂. In spite of the fact that the surface applied on the Leien of Antwerp is quite important, one has to notice the relative small width of the photocatalytic parking lane in comparison with the total street: 2*4.5 m on a total width of 60 m.



Figure 5 - Separate parking lanes at the Leien of Antwerp with photocatalytic pavement blocks.

Two different types of tests were carried out. First of all, pavement blocks were taken from the Leien after different periods of exposure. These blocks were measured in the laboratory without washing of the surface and with washing of the surface. The results are presented in Figure 6. They indicate a good durability of the efficiency towards NO_x abatement. The deposition of pollutants on the surface leads to a decrease in efficiency which can be regained after washing. To check the longevity of the photocatalytic action, measurements were recently (2010) repeated on locally removed paving stones, as shown in Figure 7. The results indicate that even after more than 6 years of service life, the durability of the photocatalytic materials still persists.

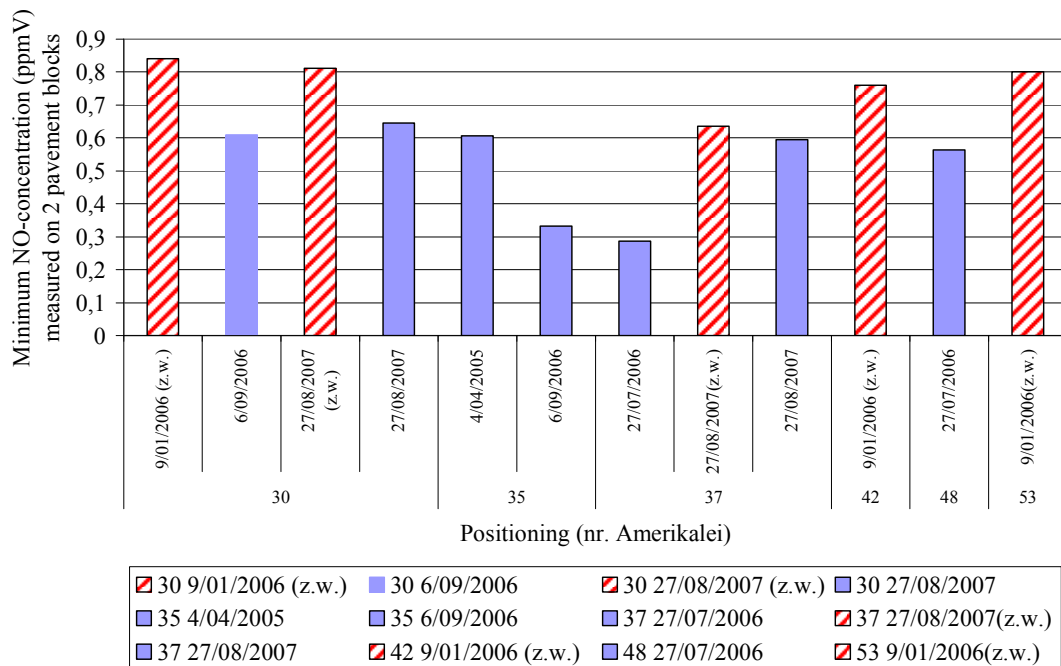


Figure 6 - NOx concentration at the outflow, measured on 2 pavement blocks, before (z.w.) and after washing the surface (purple).

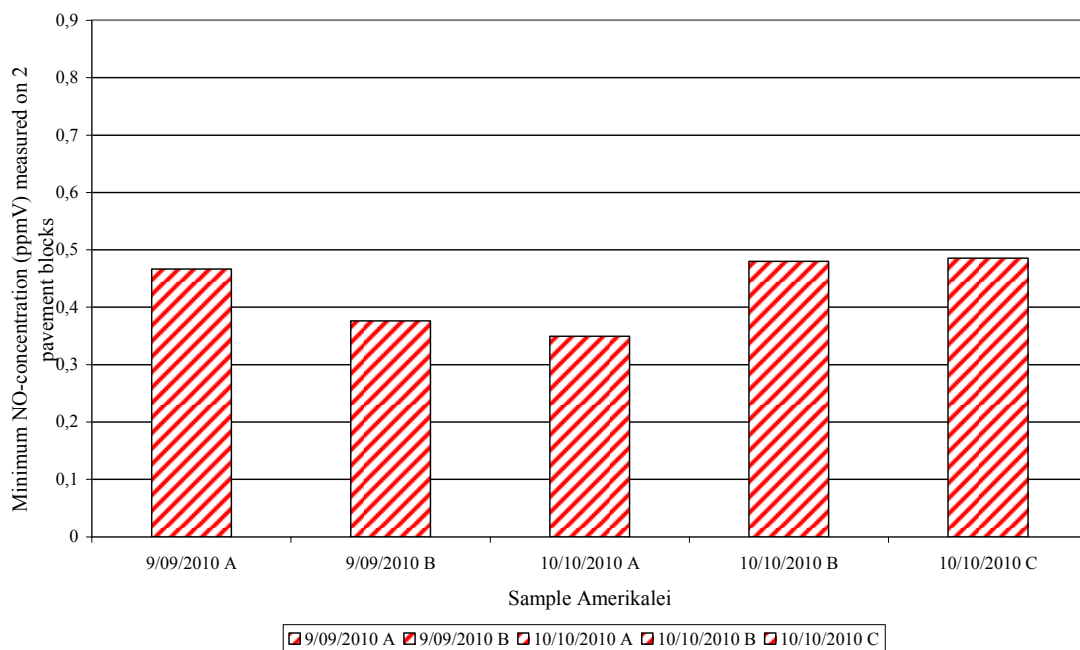


Figure 7 – Measurements on pavements blocks in 2010, after 6 years of service life.

Besides the measurements in the laboratory, on site measurements were also carried out. Since no reference measurements without photocatalytic material exist, the interpretation of these results is rather difficult. Especially the influence of traffic, wind speed, light intensity and relative humidity are playing an important role.

Figure 8 gives an overview of the measurements made on the 9th of June 2006 at 3 different places at the Leien of Antwerp. Consequently, measurements were made at house number 50, 108 and 38 of the Amerikalei. The parking lanes at number 50 and 38 contained photocatalytic pavement blocks, while the parking lane at number 108 was

placed with classic concrete pavement blocks. The measurements were carried out during 1 day. Parameters of the measurements are shown in Table 1.

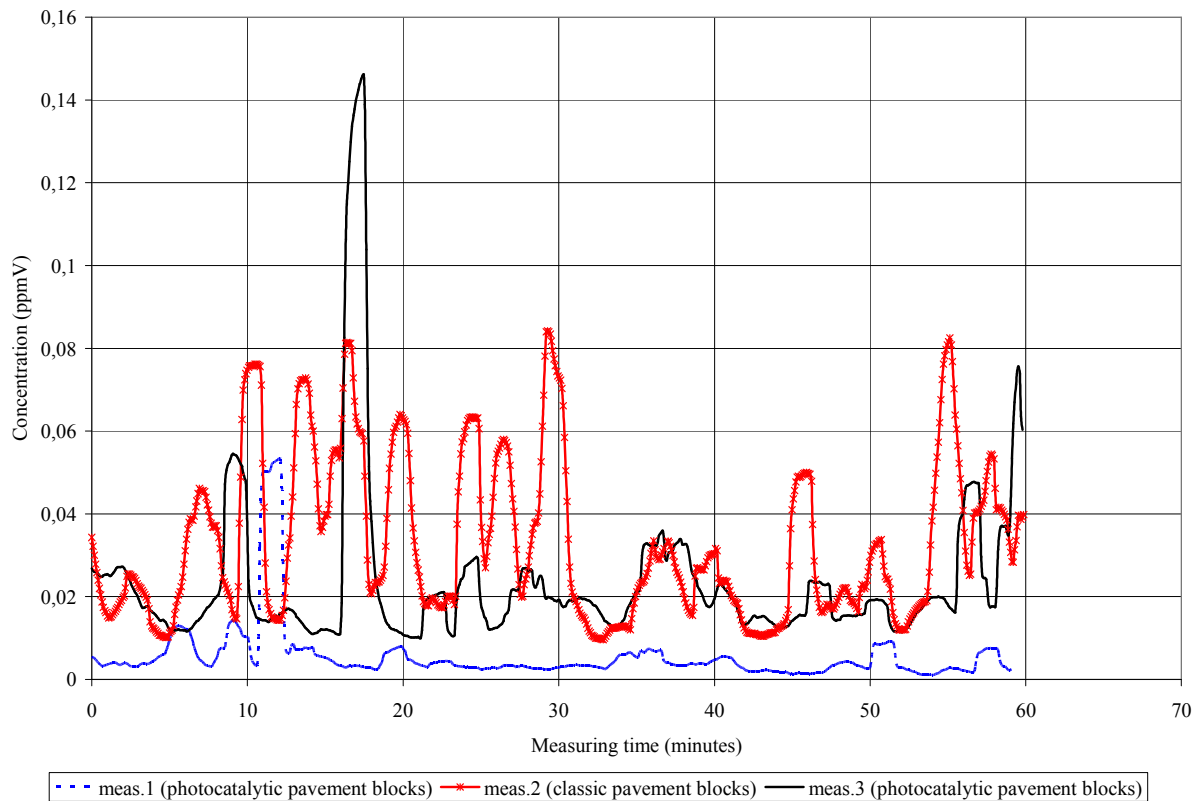


Figure 8 - NOx measurements at the Leien of Antwerp.

Table 1 - Parameters of the measurements at the park lanes of the Leien of Antwerp.

	Time	Vehicles/hour	R.H.	Temperature	Light intensity
1	10:32–11:32	1332	33 %	25°C	12 W/m ²
2	11:41-12:41	1494	30 %	26,6°C	17 W/m ²
3	14:10-15:10	1620	25 %	32,4°C	25 W/m ²

The air was continuously taken at 5 cm above the surface at the side of the photocatalytic pavement blocks. The light intensity was measured parallel to the surface of the pavement blocks. The vehicles were counted manually on the main road of the Leien of Antwerp during the last 10 minutes of the measurement.

The results indicate a decrease in NOx-concentration at the sites with photocatalytic materials. A levelling out of the peaks is visible. The 3rd measurement is slightly higher than the 1st in spite of the lower relative humidity. Although the results presented in Figure 8 give an indication of the efficiency of the photocatalytic material in situ, precaution has to be taken since the results are momentary and limited over time.

5. RECENT PHOTOCATALYTIC APPLICATIONS IN BELGIUM

Since the first application in Antwerp (2004-2005), much progress has been made within the photocatalytic research area. Newer, better and more efficient materials are constantly being developed, and action is more and more broadened also to visible light responsive materials. An example of such a material is given in Figure 9. Hence, the need still exists to develop more in situ applications in which the relation between the efficiency in laboratory and on site is established (see e.g. [12] and [13]). An overview of two such recent projects in Belgium is given in this section.

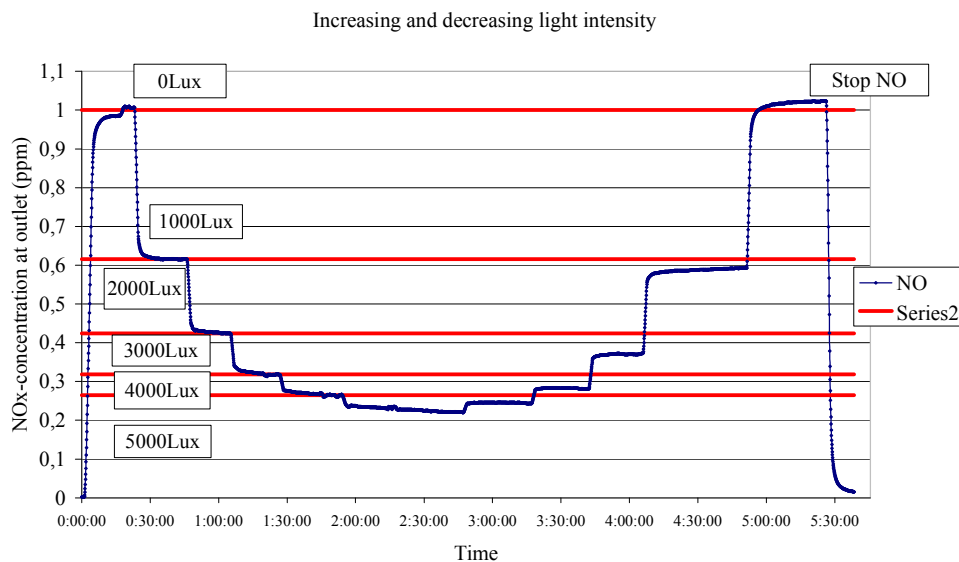


Figure 9 - NO-reduction under visible light as a function of different intensities [11].

5.1 Life⁺-project PhotoPAQ

The Life⁺-project PhotoPAQ, *Demonstration of Photocatalytic remediation Processes on Air Quality* [4], is aimed at demonstrating the usefulness of photocatalytic (road) construction materials for air purification purposes in an urban environment. Within this consortium, consisting of 7 partners coming from 5 different countries, two extensive field campaigns will be organized in Europe, of which one in Belgium. For the latter, photocatalytic cementitious materials will be applied on the side walls and roof of the Leopold II tunnel in Brussels, see Figure 10.



Photocatalytic applications in Belgium, purifying the air through the pavement

Figure 10 – Inside view of test site within Leopold II tunnel in Brussels for PhotoPaq project

A test section of about 200m in length is foreseen, which will be renovated in summer 2011. Afterwards, an intensive measurement campaign will take place in September 2011 to rigorously assess the effect on air pollution inside the tunnel. To this end, a dedicated UV-lighting system will also be installed inside the tunnel which can be modulated (on/off) to directly see the action of the photocatalytic walls. Concurrently, simulations of the tunnel air flow will be done in order to model the abatement of pollutants and the effect of different influencing parameters (traffic flow, concentration profiles, ventilation...). This modeling, when validated with measurements, could provide a valuable tool for extrapolation of the findings to other sites. First results for this project are expected in beginning of 2012.

5.2 INTERREG project ECO2PROFIT

The broad environmental sustainability project ECO2PROFIT deals with reduction of the emission of greenhouse gasses and sustainable production of energy on industrial estates in the frontier area between Flanders and Holland. To reach these goals, several tangible demonstration projects have been planned on industrial sites in Belgium and the Netherlands.

One particular project is situated on the industrial zone “Den Hoek 3” in Wijnegem (province of Antwerp). Here, the regional development agency POM Antwerp is aiming to use a double layered concrete with recycled concrete aggregates in the bottom layer, and photocatalytic materials (TiO_2) in the top layer, for the road construction. That way, air purifying and CO_2 reducing concrete roads can be built which are both innovative and energy efficient. For this project, the BRRC was asked to set-up an elaborate testing program in the lab to help optimize the air purifying performance of the top layer, without interfering with other properties of the concrete (workability, mechanical, durability...). Furthermore, a trial section was also constructed, as shown in Figure 11) to get familiar with the technique of constructing two-layered concrete.



Figure 11 – Trial section of double layered concrete with TiO_2 on the top layer in industrial zone “Den Hoek 3” in Wijnegem

For the application of photocatalytic materials in a concrete road (and in general for any other type of application) a fundamental choice can be made between: mixing in the mass (TiO_2 in cement) and/or spraying on the surface (dispersion of TiO_2). The former has the advantage of a more durable action since the TiO_2 will continuously be present, even after wearing of the top layer. On the other hand, the initial cost will be higher (higher TiO_2 content, necessity for double layered concrete) and only the TiO_2 at the surface will be active. In contrast, dispersing at the surface of a TiO_2 solution will provide a more direct action, and a lower initial cost (“ordinary” cement). In this case, however, the longevity of

Photocatalytic applications in Belgium, purifying the air through the pavement

the photocatalytic action could be questioned because of loss of adhesion to the surface in time. This fundamental choice was also investigated within the research programme.

For reasons of noise reduction and comfort of the user, it was decided to use an exposed aggregates surface finish (grain size 0/6,3) for the top layer (see for instance on the right of Figure 11). In a first phase, an optimization of the concrete composition for both layers had to be performed in terms of workability (ideal grading curve), mechanical properties (compression strength), and durability (resistance against frost/thaw cycles), as illustrated in Figure 12.

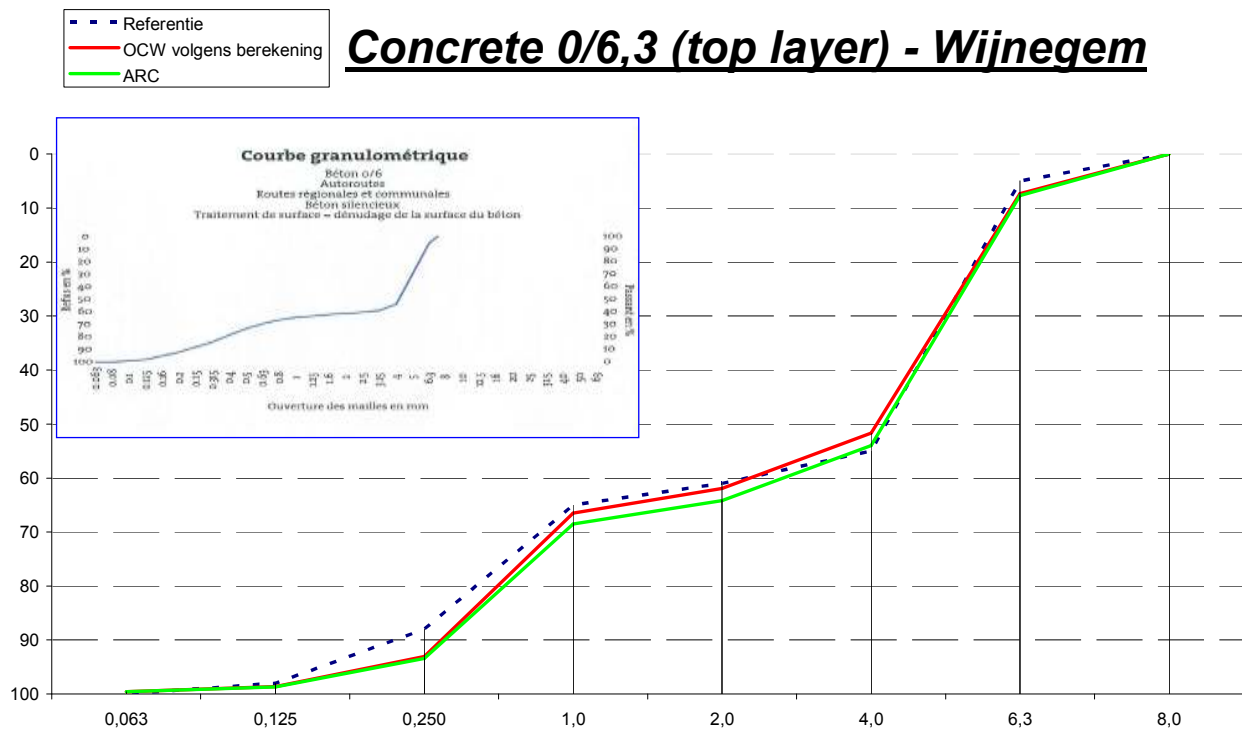


Figure 12 – Optimisation of concrete for top layer to approach the “ideal” inert skeleton.

Subsequently, trial sections of 30 m (Figure 11) were constructed to assess the feasibility of using the double layered technique in practice. In this stage, a “normal” cement was used (no mixing of TiO₂ in the mass) while a photocatalytic dispersion was sprayed over the exposed aggregate surface, after application of a curing compound. In addition, some first tests towards the air purifying performance of the concrete surface were made. To this purpose test plates were also made on site with the concrete of the top layer and with application of the photocatalytic dispersion. After construction these plates were kept under atmospheric conditions for 20 days, and subsequently cut to size and stored at RH = (60±2)% and T = (20±2)°C for 15 days. Then the samples were tested for their photocatalytic efficiency in NO_x abatement with the set-up of Figure 2 according to the ISO norm [9]. The results are shown in Figure 13.

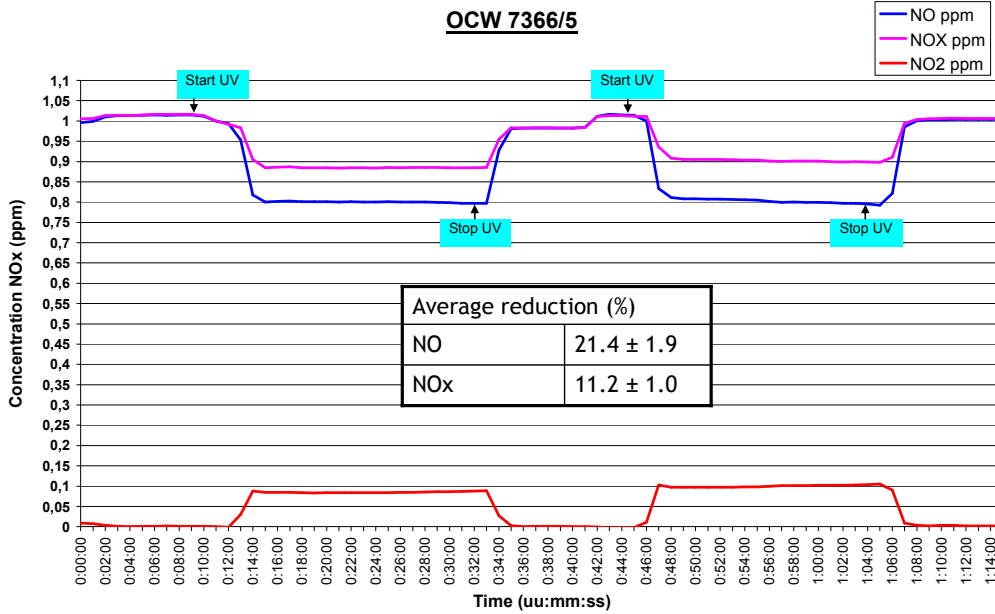


Figure 13 – Photocatalytic efficiency for test plates of trial section in Wijnegem (photocatalytic dispersion on the surface)

Figure 13 clearly shows repeatable photocatalytic efficiency for these first materials. In order to improve the air purifying performance, further testing was undertaken in the laboratory in which the effect of different, important factors was studied:

- Effect of different materials in the mass
- Effect of different dispersions on the surface
- Effect of curing compound
- Effect of curing and/or conservation conditions
- Effect of surface finishing
- Efficiency after longer time in use

First of all, it appeared that different photocatalytic materials available on the market, for mixing in the mass or applying on the surface, can give drastically different results regarding air purifying performance, as shown in Figure 14.

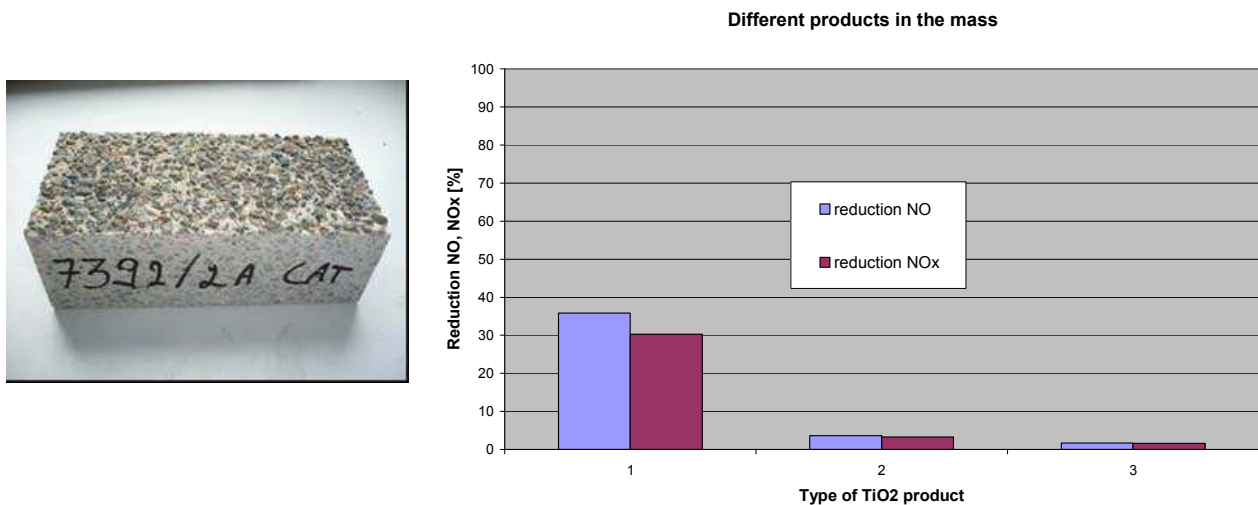


Figure 14 – Effect of different photocatalytic products mixed in the mass of the concrete (no curing compound, modified curing procedure)

A second important influencing factor is the curing compound, which is normally applied after exposing the aggregates at the surface, to protect the concrete against desiccation. This is approximately 24 hours after putting the concrete in place. The effect is illustrated in Figure 15.

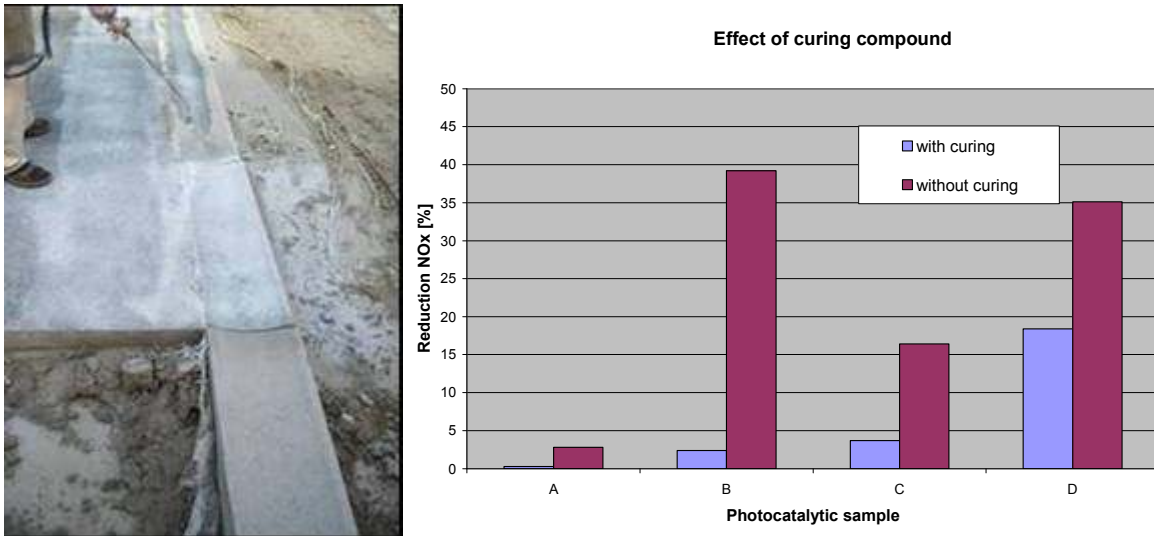


Figure 15 – Effect of curing compound on photocatalytic efficiency (different products, mass and/or dispersion, with and without curing)

Clearly, the curing compound will initially inhibit the photocatalytic reaction, probably because it is shielding off the active components from the pollutants in the air. Consequently, the curing must disappear from the surface (normally after 1-2 months open to traffic) before the TiO_2 will reach its optimal air purifying performance. In case of a TiO_2 spray, this also means that it is best to apply the photocatalytic dispersion some time after the curing compound to have the best effect.

Besides the curing compound, also the storage and curing conditions of the concrete plays a role, although to a lesser extent compared to the former. This is demonstrated in Figure 16.

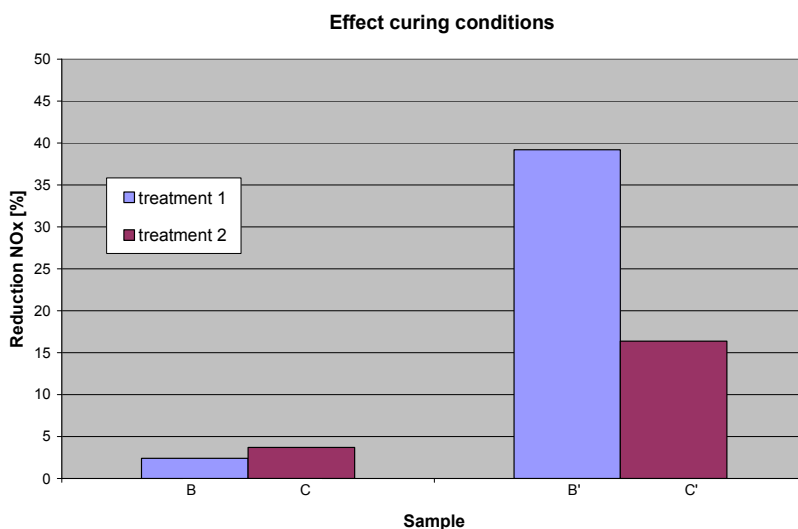


Figure 16 – Effect of curing conditions: treatment 1 = 7 days at RH>95%;T= (20±2)°C + 28 days at RH= (60±2)%;T= (20±2)°C – treatment 2 = 35 days at RH>95%;T= (20±2)°C (B,C = with curing; B',C' = without curing).

The effect is most pronounced in the case of absence of a curing compound, where it can be seen that more humid conditions (treatment 2) have an adverse effect on the photocatalytic efficiency. This is related to the relative humidity conditions at the surface of the concrete and the competition effect between water and pollutants as described above. Moreover, the hardening process of the concrete will slightly differ depending on the curing conditions which could affect the porosity of the surface, and hence also the photocatalytic action. This could be important in practice, because it is obviously hard to control these hardening conditions in situ.

To see the effect of surface treatment and more specifically of the exposed aggregates surface finish, a comparison among three different surfaces has been made: exposed aggregates, smooth (formwork side) and sawn surface, and this for one type of product. The results are depicted in Figure 17. This shows that the exposed aggregates surface perform equally well as the smooth, formwork surface, but not as good as a sawn surface. This is the result of the combined action of less cement at the surface and higher surface porosity, two competing effects which in the end yield the final efficiency shown in Figure 17.

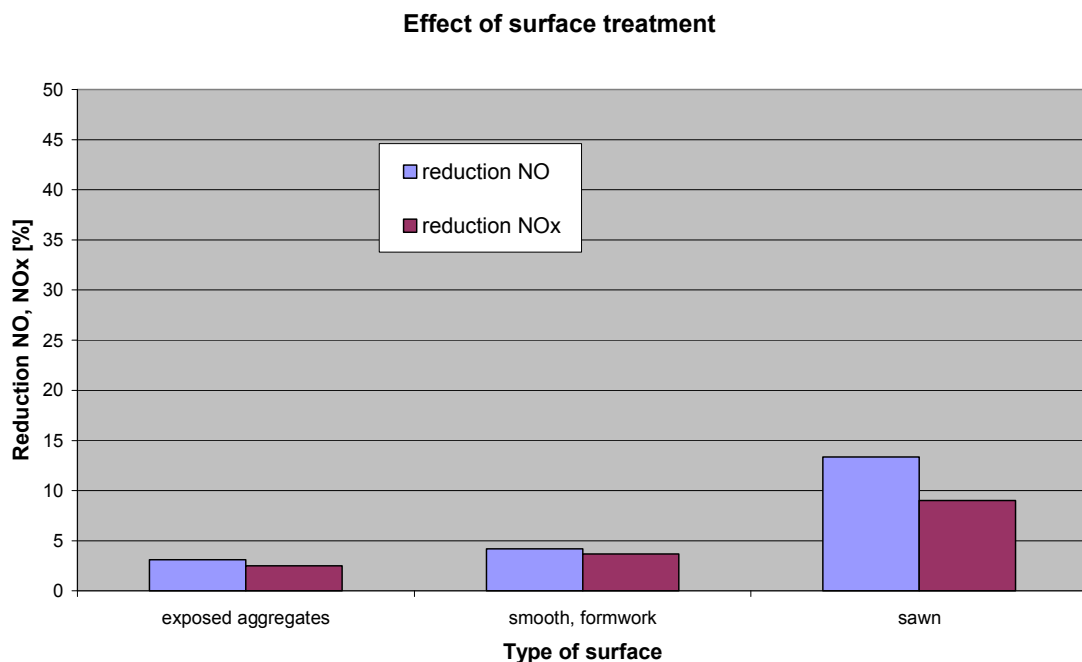


Figure 17 – Effect of surface treatment on photocatalytic efficiency (only one type of “less” active product in mass)

Finally, the durability of the photocatalytic action was also tested in laboratory by simulating the possible effect of traffic and/or weather on the surface in brushing and washing the samples. The influence of this action is illustrated in Figure 18.

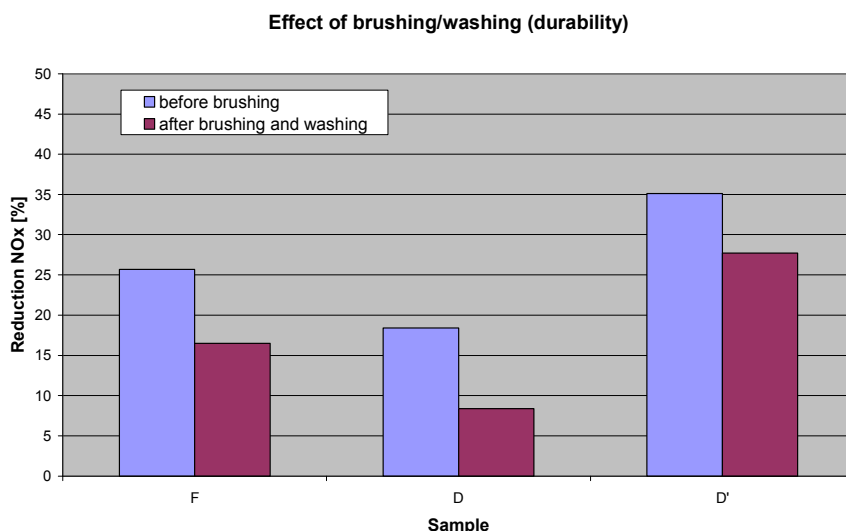


Figure 18 – Effect of brushing and washing the samples in the lab (simulation of durability)

The photocatalytic efficiency decreases by about 10% after the brushing/washing operation. This demonstrates once again the need to assess the durability of these photocatalytic materials in situ and to check to longevity of the action after several years of service life.

In conclusion, we have clearly assessed the effect of the curing compound, curing conditions and surface finish and simulated the durability of the photocatalytic action. Based on these results and the optimization of the concrete composition, a proper selection of photocatalytic materials and of application procedures could be made, for the construction of double layered, photocatalytic concrete roads on the industrial zone “Den Hoek 3” in Wijnegem. A final choice has been made to combine TiO_2 in the mass of the concrete top layer with curing compound on the surface, and spraying of a photocatalytic dispersion after 2-3 months. Furthermore, provisional controls of the photocatalytic efficiency in the lab and in situ, are planned to check the separate action of the two types of photoactive materials (mass and dispersion), and to assess the durability of the air purifying performance. Due to the hard winter in December in Belgium, the construction works can only start in March 2011, the photocatalytic efficiency will be followed afterwards (2011-2012).

6. CONCLUSIONS AND PERSPECTIVES

The use of photocatalytic materials to minimize the air pollution by traffic is applied more frequently on site in horizontal as well as in vertical applications, also in Belgium. Laboratory results indicate a good efficiency towards the abatement of NOx in the air by using photocatalytic materials. Also, the durability of the photocatalytic action remains intact. However, the relative humidity is an important parameter, which may reduce the efficiency on site. If the RH is high, the water will be adsorbed at the surface and prevent the reaction with the pollutants.

Measurements on site in the past indicated a decrease of the peaks due to the presence of the photocatalytic material. Repeated measurements in the laboratory on photocatalytic concrete pavement blocks confirm the efficiency over time, even after more than six years of service life. Although a reduction in efficiency is noticed due to the deposition of the NO_3^- on the surface, the original efficiency can be regained by washing the surface.

The translation from the laboratory results to the site efficiency is still a difficult factor, because of the great number of parameters involved. Hence, there is still a need for large scale applications to demonstrate the effectiveness of photocatalytic materials on site, including also other positive effects (O_3 , VOC's, PM...). To this purpose, two recent applications have also been started up in Belgium, which show already some promising results. In addition, the best results will be achieved by modeling the environment, validating the model by measurements followed by an implementation of the different parameters to assess the real life effect. One must bear in mind that photocatalytic applications are only effective in case of good contact between pollutants and the active surface. Parameters as wind, street configuration and pollution source play an important role.

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