ENVIRONMENTAL FRIENDLY CONCRETE PAVEMENT BLOCKS:
AIR PURIFICATION IN THE CENTRE OF ANTWERP

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SUMMARY

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

A project on environmental friendly concrete pavement blocks is conducted at the Belgian Road Research Centre, subsidised by the Flemish government (IWT). The use of photocatalytic material in the surface of pavement blocks to obtain air purifying materials is investigated. In contact with light, TiO$_2$ as photocatalyst, is able to reduce the NO and NO$_2$ content in the air, caused by the exhaust of traffic. The efficiency is tested on pavement blocks, but the technique can as well be applied on other road elements (noise reducing walls, linear elements, etc.) or as a coating on new materials or existing structures.

The first part of the project concentrated on the behaviour of the material in the laboratory. The parameters of the testing method are determined, as well as the behaviour of the material over time. The second part of the project concentrates in the translation of these laboratory results to real site measurements. A pilot project is started in Antwerp, at a main city road through the centre. 10 000 m$^2$ photocatalytic pavement blocks are placed. Measurements on the efficiency towards air purification of the pavement blocks are carried out.

This paper focuses on the environmental aspects of the types of materials, the mechanism of air purification. A review of the results obtained in the laboratory as well as first results from the site is discussed.

1. HETEROGENEOUS PHOTOCATALYSIS, A PROCESS FOR AIR PURIFICATION

1.1. Air pollution by traffic exhaust

A variety of air pollutants have known or suspected harmful effects on human health and environment. In most areas of Europe, these pollutants are principally the products of combustion from space heating, power generation or from motor vehicle traffic. Pollutants from these sources may not only prove a problem in the immediate vicinity of these sources but can travel long distances, chemically reacting in the atmosphere to produce secondary pollutants such as acid rain or ozone.
The principle pollutants emitted by vehicles are carbon monoxide, oxides of nitrogen (NOx), volatile organic compounds (VOC's) and particulates. 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 lead to the formation of 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 NOx emissions and resulting from the transport of NOx, oxidation in the air into NO3 and finally precipitation of nitrogen acid with harmful consequences for building materials (corrosion of the surface) and vegetation.

1.2. Heterogeneous photocatalysis

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 materials can be added to the surface of pavement and building materials. In combination with light, the pollutants are oxidized, due to the presence of the photocatalyst and precipitated on the surface of the material. Consequently, they are removed from the surface by the rain.

Heterogeneous photocatalysis with TiO2 as catalyst is a rapidly developing field in environmental engineering. It has a great potential to cope with the increasing pollution. The impulse of the use of TiO2 as photocatalyst was given by Fujishima and Honda in 1972 (Fujishima and Honda 1972). They discovered the hydrolysis of water in oxygen and hydrogen in the presence of light, by means of a TiO2-anode in a photochemical cell. In the eighties, organic pollution in water was decomposed by adding TiO2 under influence of UV-light. The application of TiO2 as an air purifier originated in Japan in 1996. A broad spectrum of products appeared on the market for indoor use as well as for outdoor use. Heterogeneous photocatalysis with TiO2 as catalyst results in a total mineralization of a broad gamma of organic compounds (alkanes, alkenes, alcohol, pesticides, etc.) Further, it is possible to reduce NOx, bacteria, viruses, etc. The speed at which these reactions take place depends on the intensity of the light, the environmental conditions (temperature, relative humidity), the amount of TiO2 present at the surface and the adhesion of the pollutants to the surface. In the case of traffic, 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, influence the final reduction rate of pollutants in situ.

1.3. TiO2 and the photocatalytic process

TiO2 is a metal, which is multiply present in nature. The oxygen TiO2 has three different molecule structures: rutile, anatase and brookiet (Fujishima et al. 1999). Rutile is known as pigment in white paints, but shows up till now low photocatalytic reactivity. Anatase is preferable if used as photocatalytic cell. To use anatase in heterogenic photocatalysis, UV-light with a wave length lower than 387 nm has to be present. Also the intensity of the light is important to optimize the photocatalytic activity. Normal daylight can be used for the photocatalytic reaction. Research is focusing now on the application of nano-particles of TiO2, active in the visible light range.

Existing applications may be found in water purification, air conditioning (air purification), self-cleaning glazing, ceramic tiles (self-cleaning, antibacterial), textile (anti-odour), mirrors (anti-condensation), tunnel lightning, white tents, etc. Besides the air purifying and antiseptical action, where the pollutants are oxidized or reduced due to the presence of the photocatalyst, TiO2 is also used to obtain a self-cleaning material. This is due to a very high hydrophilicity of the surface when TiO2 is activated by UV-light. The water layer is attracted between the dirt and the surface resulting in the
washing off of the dirt particles. This effect is more pronounced with smooth surfaces like glass and ceramic tiles. In the case of concrete surfaces, the self-cleaning effect will be more limited due to the physical anchoring of the dirt in the larger pores. In addition, due to photocatalytic working, a decomposition of the dirt particles, especially of the organic particles takes place, followed by the washing of the surface resulting in a cleaner surface.

1.4. Application of TiO₂ in building materials as photocatalyst
The increase in patents during the last decade indicates a huge interest, especially from Japan and Europe, in the application of TiO₂ as photocatalyst in building materials. Regarding the reduction of air pollution due to traffic in urban areas, the application on pavement surfaces or on the building surfaces in cementitious materials gives optimal solutions. To increase the efficiency of the photocatalyst, its presence at the surface of the material is crucial. It has to be accessible by sunlight to be activated. Consequently, the pollutant has to be absorbed on the surface and oxidized or reduced to a less harmful element.

The goal is to have as much TiO₂ as possible at the surface of the material, without the risk of loosing it by abrasion or weathering. Up till now, the most efficient way to apply the TiO₂ is in a thin layer cementitious material, which is placed on the surface. Application in concrete tiles is therefore very suitable: the TiO₂ can be added to the weathering layer. If the layer is slightly used, new TiO₂-particles will be present at the surface.

Other applications can be found in architectural concrete. The use of white cement with TiO₂ at the surface of buildings and construction attribute to the durability of the visual aspect of the building. Due to the photocatalytic action, the whiteness of the building will remain and dirt will be washed away more easily due to the hydrophilic properties or will be decomposed. The application of photocatalytic panels at the facades of buildings is investigated in the European PICADA project (Maggos et al. 2005).

2. LABORATORY RESULTS: PARAMETER EVALUATION
To determine the air purifying activity of TiO₂, applied in building materials, the oxidation of NO and NO₂ into NO₃ is determined. Emphasis is put on this pollutant, since it is one of the most important pollutants produced by traffic and plays a major role in the formation of smog and ozone. It is also one of the pollutants on which limits have been placed by the Kyoto agreement. The oxidation of the NO is simplified presented by the following equations:

\[ \text{NO} + \cdot \text{OH} \xrightarrow{h\nu, \text{TiO}_2} \text{NO}_2 + \text{H}^+ \]

\[ \text{NO}_2 + \cdot \text{OH} \xrightarrow{h\nu, \text{TiO}_2} \text{NO}_3^{-} + \text{H}^+ \]

Other parameters which are important are the relative humidity and the temperature. At higher temperatures, the conversion will be better. The relative humidity is important, since the water in the atmosphere plays a role in the adhesion of the pollutants at the surface and therefore also the conversion rate. In the case of a higher relative humidity, the conversion will be lower. Optimal conditions are therefore reached on hot summer days with high temperatures and low R.H. It is also on those days that the risk on smog during the summer is the highest and thus the efficiency of the air purification is the highest.
The NO$_3^-$, which is formed during the process, will precipitate on the surface of the stone. To retain the efficiency of the material, the deposit will have to be washed away by rain or by cleaning the surface with water.

3.1. Test set-up
The test set-up is based on the Japanese standard JIS TR Z 0018 “Photocatalytic materials – Air purification test procedure”, which is also adapted in a proposal for an ISO standard (2/2/2004): “Fine ceramics (advanced ceramics, advanced technical ceramics) – Test method for air purification performance of photocatalytic materials – Part 1: Removal of nitric oxide”, ISO TC 206/SC N. The test set-up consists of a metal container, in which 1 pavement block is placed, with a UV-transparent glass at the top. Air with a NO-concentration of 1 ppmV is blown over the surface with a flow rate of 3 l/min. The height of the free space is 3 mm. The temperature is approximately 23°C and the relative humidity is 50%. The light intensity is equal to 10 W/m$^2$ in the range between 300 and 460 nm. The maximum is at 365 nm. The set-up is illustrated in Figure 1.

Figure 1. Set-up of the test to determine the air purifying properties with large container (3*2 rows of pavement blocks)

The concentration of NO and NO$_2$ is measured at the outlet of the container. The test is executed during 5 hours. 30 minutes prior to the test, the concentration of the air is measured without illumination to ensure that there is no deposit of the NO on the surface.

3.2. Results
A typical result of the test is given in Figure 2. The inlet concentration is equal to 1 ppmV. As soon as the light is put on, the concentration drops with approximately 40%, depending on the type of material. After 5 hours of illumination, the NO is cut off for 30 min. Consequently, the light is put off again and the NO is concentration is measured. The results indicate a small increase in NO$_2$, but a significant decrease in NO$_x$ (NO+NO$_2$). The final decrease depends on the material itself, on the size of the surface exposed, on the concentration of NO, on the light intensity, the ambient temperature and the flow rate.
By increasing the surface by 6 (2 rows of 3 pavement blocks) for instance, a further reduction is obtained (up till 85%), which results in a NO$_x$ concentration of 13.5% at the outlet, as can be seen in Figure 3. This is very promising for the extrapolation to the situation in situ. By increasing the time of contact or increasing the surface over which the air flows, the reduction will be even more significant.

The laboratory research program consisted of the control of different parameters such as temperature, relative humidity, contact time (surface, flow velocity, height of the air flow over the sample, etc.). In general it can be stated that the efficiency towards the reduction of NO$_x$ increases with a longer contact time (larger surface, lower velocity, smaller height of air flow), with a higher temperature and lower
relative humidity. 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 present.

Figure 4. Influence of the relative humidity on the efficiency of photocatalytic pavement blocks

Figure 4 gives an idea of the influence of the relative humidity on the efficiency of photocatalytic pavement blocks. The influence depends to a large extend on the type of material used. Apparently, the hydrophilic effect at the surface is gaining over the oxidizing effect. The water molecules are adsorbed at the surface and prevent the pollutants to reach the TiO₂. If the relative humidity decreases again, the efficiency increases.

4. PILOT PROJECT IN ANTWERP

An important issue is the conversion of the results, obtained in the laboratory to real applications. The construction of a test section of 10,000 m² photocatalytic pavement blocks as pilot project on the parking lanes of a main axe in Antwerp allows us to search for answers on frequently asked question, like the amount of reduction of NOₓ, the durability of the material not only for mechanical and aesthetical properties, but also for the photocatalytic efficiency, the minimum surface needed related to traffic density and the frequency of maintenance, i.e. washing off of the surface by rain or by artificial spraying.

Figure 5 gives a view of the parking lane, where the photocatalytic concrete pavement blocks are applied. Only the upper layer of the blocks contain TiO₂.
Figure 5. Separate parking lanes at the Leien of Antwerp with photocatalytic pavement blocks

To obtain results towards the reduction of e.g. NO\textsubscript{x} and on the durability of this reduction, different measurement techniques are applied. The use of the global measurement sites is not appropriate, due to the limited surface covered by the photocatalytic pavement blocks, compared to the overall surface. Local continuous measurement of the NO and NO\textsubscript{2} concentration demands a long period during which the measurement has to be conducted, to eliminate the influence of parameters such as wind, temperature, light intensity, traffic intensity and so on. Although the presence of a reference lane allows comparing measurements, the time needed to come to reliable results is very long. Nevertheless, measurements during several hours on sunny days are carried out. Another method, which seems to be more suitable, is the measurement of NO\textsubscript{3}-deposite on the surface of the blocks. The NO and NO\textsubscript{2} which is oxidized into NO\textsubscript{3} is deposited on the surface. By washing off the stones with distilled water and determining the amount of N in the water, an idea of the minimum amount of reduced NO\textsubscript{x} can be obtained. Furthermore, a regular measurement in the laboratory is executed on blocks taken from the test site to measure the possible reduction of efficiency under controlled conditions.

The laboratory tests are made on 3*2 pavement blocks, taken from the road in Antwerp. The average results are presented in Table 1. Reading the table, one should keep in mind that the results presented here are for 2 pavement blocks and therefore not directly transferable to the real site. Not only the surface and the flow are not comparable, but also the initial NO-concentration is much higher than in situ. On the other hand, they give an indication of the durability of the photocatalytic effect.

<table>
<thead>
<tr>
<th>Time</th>
<th>NO-concentration after passing over the pavement block</th>
<th>NO\textsubscript{2}-concentration</th>
<th>NO\textsubscript{x}-concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>April 2005</td>
<td>0.60 ppmV</td>
<td>0.01 ppmV</td>
<td>0.61 ppmV</td>
</tr>
<tr>
<td>January 2006</td>
<td>0.80 ppmV</td>
<td>0.01 ppmV</td>
<td>0.81 ppmV</td>
</tr>
<tr>
<td>June 2006</td>
<td>0.75 ppmV</td>
<td>0 ppmV</td>
<td>0.75 ppmV</td>
</tr>
</tbody>
</table>

The results indicated in Table 1 show indeed a decrease in efficiency, which seems to stabilize over time. The measurements will continue over 2 more years to see the influence of aging, dirt, season, etc.
5. CONCLUSIONS AND FURTHER RESEARCH

The addition of TiO$_2$ in building materials adds an additional property to the road. Purification of the air, which is in contact with the surface, is obtained when the surface is exposed to UV-light (present in daylight). The measurements in the laboratory on photocatalytic pavement blocks gave good results towards air purification, measured as NO$_x$ reduction. The best results were obtained by high temperature (> 25°C), low relative humidity, high light intensities and long contact times. This situation is obtained on hot sunny days, without any wind, when the risk on smog formation due to the high rate of pollution is the highest.

A pilot project of 10,000 m$^2$ is constructed in Antwerp. Measurements to reveal the air purifying efficiency in situ and the durability of this efficiency are programmed. A reduction of 20% of purification efficiency is measured after 1 year in service.

6. REFERENCES


7. ACKNOWLEDGEMENTS

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