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# Durability of epoxy-bonded TiO<sub>2</sub>-modified aggregate as a photocatalytic coating layer for asphalt pavement under vehicle tire polishing

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#### ABSTRACT

A new approach to construct photocatalytic air-purifying asphalt pavements coated with  $TiO_2$  modified aggregate was developed and its feasibility and performance were evaluated. Two methods, namely the surface coating and pore filling methods, were adopted to produce the  $TiO_2$  modified aggregate. Their photocatalytic efficiency and mechanical performance under vehicle tire polishing, applied by an Aachen Polishing Machine (APM), were investigated and compared. Results indicated that it is feasible to build durable photocatalytic pavement using the new approach and material spreading process reported in this study. Both  $TiO_2$  modification methods provided the spreading aggregate with excellent and comparable NO degradation rates before polishing; however, the pore filling method provided better long-term NO degradation efficiency. The spreading aggregate modified by either method showed excellent long-term skid resistance and surface texture properties after vehicle tire polishing.

#### 1. Introduction

Roadside air pollution caused by automobile exhausts is a serious environmental concern, especially in high population-density cities. Nitrogen oxides (NO<sub>X</sub>), one of the most hazardous components in vehicle emission, are harmful to both the atmospheric environment and human health. Conventionally, various measures such as using cleaner fuel and installing gas-cleaning equipment in vehicle exhaust systems have been implemented to counteract this issue by reducing the emissions from vehicle. Recently, studies have been conducted on photocatalytic pavements which can assist in the degradation of NOx into harmless substances [1-3]. The air-purifying function of photocatalytic pavements is achieved by incorporating or coating the pavement with catalysts, in most cases titanium dioxide (TiO<sub>2</sub>), which are capable of degrading nitric oxide (NO) and nitrogen dioxide (NO<sub>2</sub>) under ultraviolet (UV) irradiation [2-5]. Before being applied in photocatalytic pavement, TiO<sub>2</sub> had been used as a self-cleansing material in many fields due to its air purifying function [6,7]. Under UV irradiation, TiO<sub>2</sub> generates powerful oxidizing agents, which have the strong capability of oxidizing NO<sub>X</sub> to nitric acid (HNO<sub>3</sub>), which is the final product of the degradation process. Trace amounts of HNO3 have limited effects on the pavement performance, and can be easily washed away by rainwater [4,6,7].

To achieve satisfactory air-purifying performance,  $TiO_2$  particles should be exposed to UV light and in direct contact with  $NO_X$ pollutants. Correspondingly, it is important for the coating materials of photocatalytic pavements to have sufficient contact area among  $TiO_2$ particles, pollutants and sunlight and to have sufficient resistance against vehicle tire polishing. In addition, it is necessary to ensure that the incorporation of  $TiO_2$  particles does not compromise the original mechanical performance of pavements, such as skid resistance and other mechanical properties related to wear due to traffic.

Currently, different methods have been attempted to apply  $TiO_2$  particles onto both asphalt and concrete pavements, such as: a) mixing TiO2 with water solution or asphalt emulsion and then spraying them onto road surface [3,8], b) Using nano- $TiO_2$  particles as asphalt modifier [9–11], c) incorporating  $TiO_2$  to crumb rubber surface and then spraying the  $TiO_2$ -crumb rubber mixture onto pavement surface during the construction process [12], and d) coating asphalt pavement surface with asphalt emulsion containing micro pores embedded with nano-TiO2 particles [13]. However, so far none of abovementioned methods provide sufficient  $NO_X$  removal efficiency and durability allowing for a wider practical application.

In Germany, the process of bonding spreading aggregate to pavement surfaces with epoxy has been used as an effective surface treatment method to enhance smoothness, improve friction, and reduce

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Fig. 1. Surface treatment of asphalt pavement with epoxy-bound spreading material.

tyre-road noise of pavements (Fig. 1) [14]. Inspired by this treatment method, this study aims to develop a new method to construct photocatalytic pavements by incorporating TiO<sub>2</sub> particles into the spreading aggregate. As this form of modification consists of applying TiO<sub>2</sub> modified material onto existing pavement surfaces, it is of upmost importance to systematically evaluate the durability of spreading materials in terms of both photocatalytic functionality and mechanical properties under tire polishing. To achieve this objective, TiO<sub>2</sub> modified aggregates were prepared with two different processes, namely the surface coating method and the void filling method. The TiO<sub>2</sub> modified aggregates were then bonded onto asphalt pavement surface to provide the photocatalytic functionality in addition to other functions, such as skid resistance. Since there is no direct contact between TiO<sub>2</sub> and asphalt in these two methods, the asphalt surface is protected from the photocatalytic processes. Moreover, high durability of photocatalytic function is expected, because TiO<sub>2</sub> particles in the aggregate may keep rising even the aggregate particles are polished. To quantify the durability and polishing resistance of the photocatalytic TiO<sub>2</sub> modified aggregate layer, a unique custom-designed Aachen Polishing Machine (APM) was applied to simulate the polishing effect of vehicle tires in the laboratory. Both the NOx removal efficiencies and the skid resistances of the photocatalytic pavements prepared with the new approach before and after APM polishing were measured to evaluate and compare the performances of two methods to modify aggregate with TiO<sub>2</sub>.

#### 2. Materials and testing program

#### 2.1. Photocatalyst

The photocatalytic efficiency of  $\text{TiO}_2$  containing materials is dependent on various factors, such as  $\text{TiO}_2$  content, UV irradiation intensity, and  $\text{TiO}_2$  type. In this study, an anatase type  $\text{TiO}_2$ , labelled as VU7, was used, because previous studies have shown that VU7 provided the highest NO-decomposition rate among various common types of  $\text{TiO}_2$  available in Germany [15,16]. Table 1 shows the basic properties of VU7.

#### 2.2. Test sample preparation

#### 2.2.1. Preparation of $TiO_2$ modified aggregate

In this study, the photocatalytic coating layer was prepared by spreading  $TiO_2$ -modified aggregate onto a thin layer of epoxy resin at asphalt pavement surface. As Fig. 2 shows, two methods were adopted to prepare the  $TiO_2$  modified aggregate, namely the surface coating method and pore filling method. In the surface coating method, the neat aggregate is mixed with cement, water and 4 M%  $TiO_2$  in a rotating drum, leading to aggregate coated with  $TiO_2$ -cement film. In the pore filling method, a porous aggregate, basalt lava, which has a void content of approximately 25 vol%, was used. The basal lava aggregate

Table 1Basic properties of the selected TiO2.

Label	Mineralogical type	Grain size (nm)	Surface area (BET) (m <sup>2</sup> /g)	рН	TiO <sub>2</sub> content (M%)
VU7	Anatase	15	90	1.5	99

was first submerged in a 4 M% TiO<sub>2</sub> cement suspension below atmospheric pressure for one hour to allow TiO<sub>2</sub> cement to penetrate into the surface pores of the aggregate. Before complete curing of the cement mortar, the excessive cement was manually removed from the aggregate surfaces by means of brushes, after which the aggregate was oven dried at 105 °C to constant mass.

The different methods underwent systematic investigations with regard to their mechanical strength, polishing resistance and wear/ abrasion resistance, confirming that the modified aggregate complies with the respective requirements as shown in Table 2. The impact crushing tests applies a defined crushing energy onto the unbound aggregate, after which the sample is passed through five sieves with defined mesh sizes. The percentage value remaining on each mesh is calculated, and the impact crushing value is calculated as the average value remaining on each mesh. The impact crushing value should be below the threshold value of 18. The polished stone value (PSV) represents the polishing resistance of aggregate and is required to be higher than 51. The chipping due to freeze-thaw-cycles (FTCs) represents the resistance of aggregate towards freeze-thaw-cycles. After ten FTCs, the mass loss is recorded and is required to be below 1 M% (F<sub>1</sub>).

#### 2.2.2. Coating asphalt pavement with $TiO_2$ modified aggregate

After the TiO<sub>2</sub> modified aggregates were prepared, they were bonded onto asphalt pavement surface with epoxy as a coating layer. A test section was designed and constructed at the Institute of Highway Engineering at Aachen, Germany, with a small-scale paver and a rolling compactor, as shown in Fig. 3. The surface of the asphalt pavement was first coated with epoxy resin, and then the TiO<sub>2</sub>-modified aggregate, 2–5 mm in size, was spread and compacted on top of it. The amount of epoxy was controlled so that it could embed half of the diameter of the largest TiO<sub>2</sub>-modified aggregate. Finally, the excessive loose aggregate was cleaned using a broom after the epoxy resin hardened. The whole process of coating asphalt pavement with TiO<sub>2</sub> modified aggregate is illustrated in Fig. 4.

### 2.3. Testing program

#### 2.3.1. $NO_x$ degradation efficiency

To measure the NO<sub>x</sub> degradation efficiency, the custom-designed testing system as shown in Fig. 5 was used, and the dimensions of the testing samples were  $10 \text{ cm} \times 5 \text{ cm} \times 1 \text{ cm}$  [17]. As aforementioned, the NOx degradation efficiency of TiO<sub>2</sub> is dependent on the test conditions and might be affected by various factors. For example, one previous study which used the same type of TiO<sub>2</sub>, VU7, in paving blocks investigated the effects of UV intensity, wind speed, and humidity on the NOx degradation efficiency of the paving blocks [18]. It was found that the NOx degradation efficiency increases with the increase of UV intensity and wind speed. In general, higher humidity led to lower NOx degradation efficiency. But the decrease in NOx degradation efficiency was not significant when the humidity was lower than 60%, and became significantly faster when the humidity was above 60%. As a result, it was recommended to control the humidity for laboratory NOx degradation efficiency tests within the range of 50% to 60%. To ensure that the NOx degradation efficiency test results conducted by different researchers are comparable, ISO 22197-1 has been developed, which specifies the standard test conditions and procedure, which have been followed in this study as described in the following paragraphs.

All test samples were first cleaned by a brush with water. Then they were slowly shaken in ultra-high quality (UHQ) water for 1 h while being irradiated at an intensity of  $700 \text{ W/m}^2$ . After that, they were dried at 60 °C for 1 h, and stored in a dehydrator for testing.

A xenon-lamp was used to irradiate the sample placed in the testing chamber. As Fig. 6 shows, the radiant energy was  $304 \text{ W/m}^2$  within the wavelength range relevant to the photocatalysis (300-400 nm). Such radiation intensity was selected because it is close to the sun radiation intensity at mid-latitudes. When the test started, a moisturized mixture

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