Functional lightweight concrete: aiming at an improved indoor air quality

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Abstract

In this research, a functional lightweight concrete is developed, with the advanced functionality of indoor air purification. Heterogeneous photocatalysis is applied and a visible-light responsive photocatalyst is incorporated in the concrete design. An absorptive additive, zeolite, is applied to in order to investigate its pollutants absorption ability. The PCO efficiency of the developed concrete was initially evaluated following ISO 22197-1 using a plug flow reactor, and results show excellent photocatalytic efficiencies, up to about 60% NO_x removal ability.

Furthermore, the effect of the indoor environmental parameters such as air flow rates and initial air pollutants concentration on the air pollutants removal ability is investigated. Results show that ventilation rate plays a crucial role in the air pollutants removal efficiency. Under a more realistic condition, i.e. with a flow rate of 1.5 L/min and an initial concentration of 500 ppb, a NOx removal of 94% is achieved.

Moreover, a tank reactor with a scaled volume of 45 l equipped with visible light to simulate a realistic indoor environment is employed to evaluate the performance of the developed concrete under indoor air conditions. The very good results indicate a promising potential of this functional concrete for indoor air quality improvement.

Keywords: air quality, lightweight concrete, mix design, NO_x, plug flow reactor, tank reactor

1. Introduction

Indoor air quality issue has received much attention because of the very important role it plays on human health. Nitric oxides (NO and NO_2), sulfur dioxide (SO_2) and Volatile organic compounds (VOCs), as typical inorganic and organic indoor air pollutants, can be emitted from traffic emissions from outside of buildings, cooking, combustion, exhaust gases, tobacco smoke, furniture, building materials, and can cause serious environmental problems such as acid rain and health problem like drowsiness, headache, sore throat, and mental fatigue [1]. Among other existing techniques, photocatalytic oxidation (PCO) technology is considered as one of the potentially very efficient advanced oxidation processes for the improvement of the air quality by degrading inorganic and organic air pollutants [2-5]. The most used photocatalyst - titanium dioxide (TiO_2) (pure or modified) - has received considerable attention during the past decade due to its high PCO activity, high chemical stability, low toxicity and low cost [6,7]. Under the appropriate irradiation of photon energy, equal to/higher than its band gap energy, electron-hole pairs are generated on the TiO_2. The photo-generated holes in the valence band and the electrons in the conduction band diffuse to the TiO_2 surface and they produce highly energetic hydroxyl radicals (OH•) and super-oxide radical anions ($O2•^-$), which can oxidize the adsorbed pollutants on the TiO_2 surface [8].

Recently published work on the application of TiO_2 in concrete [2,9-13] showed that this technology can be perfectly applied to building materials, such as concrete. It can be concluded that the application of finer photocatalytic materials, for instance, in a form of well-dispersed suspension leads to higher degradation

[11]. To further improve the photocatalytic efficiency and the durability, application of (meso-/micro-) porous materials can be beneficial as well [6]. In the previous research [14,15], lightweight concretes with different performances (structural lightweight concrete [14] and thermal insulating concrete [15, 16]) was developed with excellent properties and they can be directly applied as a substrate for applying the photocatalysts.

The present work aims to design an air purifying lightweight concrete. A visible-light responsive photocatalyst is introduced to concrete in a slurry form in order to achieve a better dispersion. Reference samples without light-weight aggregates incorporation are produced for comparative purpose. Lightweight aggregates are applied with different proportions to study their effect. Zeolite is applied as an adsorption additive to further improve the photocatalytic efficiency. The air pollutants removal efficiency of the developed materials is firstly evaluated through photocatalytic oxidation experiments following ISO 22197-1. Then the effect of environmental parameters such as air flow rate and initial pollutants are investigated. Furthermore, a tank reactor with a much large volume compared to the ISO reactor under indoor air condition (e.g. visible light irradiation) is employed to research the air purification performance under realistic indoor environment.

2. Experimental

2.1 Materials

The cement used in this study is CEM I 52.5 N, provided by ENCI HeidelbergCement (the Netherlands). The lightweight aggregates used here are commercially available product manufactured from recycled glass. These LWA contain a number of air pores (cellular structure) encapsulated in rather closed and impermeable outer shells, as can be seen in Figure 1. The LWA have very low particle densities (between 300 and 540 kg/m^3), which provide a great freedom for the design of lightweight concrete with desired low density. A zeolite with a density of 2.29 g/cm^3 is applied to adjust the absorption property of the designed samples. A polycarboxylic ether-based superplasticizer (BASF) is used to adjust the workability. The KRONOClean 7404 suspension (KC), containing carbon-doped titanium dioxide (C-TiO_2), was used as the photocatalyst. The KC consists of 40% of C-TiO_2, has a pH of 7-8 and a density of about 1.4 g/cm^3 (at 20°C). The particle size distribution (PSD) of the materials was characterized by Mastersizer 2000 (Malvern) (in case of powders) applying light diffraction or by traditional sieving technique (in case of aggregates).

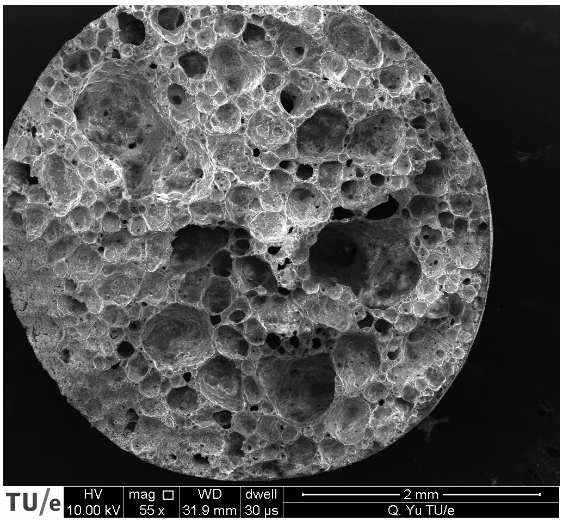

Figure 1: SEM picture of the applied lightweight aggregates.

2.2 Mix design

Six recipes containing photocatalysts were designed with or without the addition of zeolites or lightweight aggregates. Samples without addition of light-weight aggregates were used as reference. Two samples of each design were cast. The compositions of the designs are presented in the Table 1, with a SP dosage of 0.5% by mass of the applied powders.

Mix	CEM	H_2O	Sand	LWA	Zeolite	KC
1	488.76	214.81	1466.26	0	0	61.1
2			1173.01	49.99		
3			879.76	99.96		
4	474.09		1466.26	0	14.67	
5			1173.01	49.99		
6			879.76	99.96		

Table 1: Mix designs compositions [kg/m^3].

The samples were prepared, molded and cured following standard concrete preparation procedures, with the exception of addition of KC or zeolite. KC suspension was firstly mixed with the H_2O and then introduced to the system in order to get a homogenous dispersion. Zeolite was first blended with the cement to get a homogeneous dispersion. The concrete was produced using a planetary concrete mixer and the concrete was vibrated for about 30 s using a vibration table. Then the samples were covered with a plastic foil to prevent moisture loss. Subsequently, the samples were stripped from molds after 24 hours from casting, and stored in a climate chamber with a relative humidity of over 95%, at room temperature (~20℃), followi ng EN 12390-2:2000 [17], until the test age was reached.

2.3 Air pollutants removal assessment

The air pollutants removal capacity of the developed materials are assessed by using nitric oxide (NO) as a model pollutant. A plug-flow experimental setup following ISO 22197-1 is used in this study for evaluating the photocatalytic efficiency. Nitric oxide (NO) was mixed with a synthetic air and adjusted to the desired concentration and flow rate by the mass control meters. The applied light source was composed of three fluorescent tubes of 25 W each, emitting high-concentrated UV-A radiation in the range of 300-400 nm. The experimental conditions, such as the pollutant concentration, flow rate, humidity and light intensity were fully controlled and monitored. The temperature and the humidity were measured at the inlet of the reactor. The outlet concentration of NO and NO_2 were measured and interpreted as the NO_x concentration. The pollutant concentration was measured by an online NOx analyzer APNA-370 (Horiba). The APNA-370 continuously monitors the NO_x concentration using a cross-flow modulated semi decompression chemiluminescence method. The concentration measurement was performed automatically every 5 seconds with a sampling flow rate of 0.8 L/min. The active sample size was fixed as 190 x 87 mm^2.

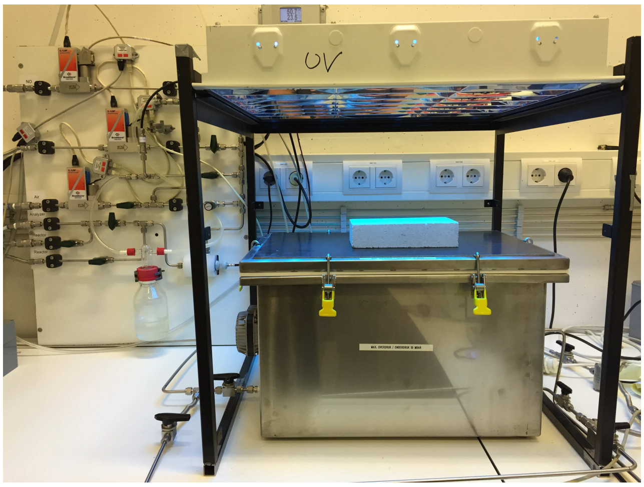

Figure 2: the employed tank reactor in the present study.

Furthermore, another reactor with different geometry is also used in this study to investigate the influence of the reactor condition. This reactor, as shown in Figure 2, is a tank reactor (, depending on the requirement fans in the reactor can be used to mix the air.

The experiments were firstly performed following the conditions set in ISO 22197-1 (denoted as ISO in Table 2). In order to further assess the performance of the developed concrete a more realistic condition, such as lower flow rate or lower pollutant concentration were also applied (see UV1-3 in Table 2). The best performing sample under the ISO standard was also selected for testing under indoor conditions (i.e. visible light irradiation, low pollutant concentration and low flow rate) (see VIS1 in Table 2).

The relevant reaction mechanism of PCO of NO_x are briefly described here [8]. Firstly, the photocatalyst needs to be photo-activated by the light of appropriate wavelength, which will result in the generation of electron/hole pairs. This is followed by the adsorption of the reactants onto the surface of the photocatalyst. The adsorbed H_2O and O_2 by the samples from the ambience leads to the trapping of the generated holes and electrons and consequent generation of hydroxyl radicals and superoxides. Afterwards, the adsorbed pollutants can be oxidized by these radicals. Due to the lack of sufficient acidic environment and the superoxide generation in the conduction band (NO is adsorbed in the valence band), it is very difficult for the superoxide to undergo the reaction. Therefore, it can be seen that NO is mainly oxidized by the produced radicals firstly to NO_2 and finally to NO_3^-.

	Plug-flow reactor					Tank reactor
Test series	ISO	UV1	UV2	UV3	VIS1	
Light source	UV	UV	UV	UV	VIS	VIS
Flow rate [L/min]	3.0	1.5	3.0	1.5	1.5	1.5
NO [ppm]	1.0	1.0	0.5	0.5	0.5	0.5
RH [%]	50					50
T [℃]	20±1					20±1
Light intensity [W/m^2]	10					10
Duration under light	1 [h]					3 [h]

Table 2: PCO experimental conditions.

The amount of NO_x ($NO+NO_2$) removed was calculated using the following equation:

$$NO_{x,con}(\%) = (C_{NO_x,in} - C_{NO_x,out}) \times 100\% / C_{NO_x,in}$$

where the initial concentration NOx was taken as an average value of 5 minutes before turning on the light. The outlet concentration NOx was taken as an average value of the last 5 minutes of the irradiation period.

3. Results

3.1 Characterization

The PSDs of the applied materials are presented in Figure 3. The average particle size of the zeolite was measured, namely 48 µm. The average particle size of the KRONOClean 7404 was measured about 150 nm. The observed particle sizes might lead to incorporation of the nano photocatalytic particles into the mesoporous system of the zeolite leading to the protection of the particles and the improved photocatalytic efficiency.

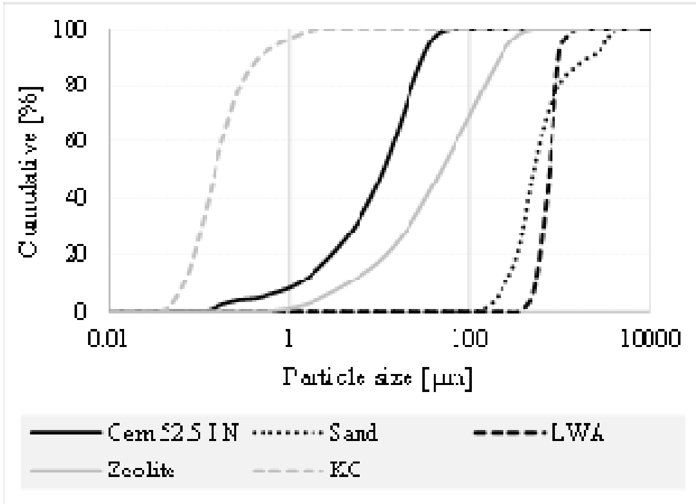

Figure 3: PSDs of the used materials.

3.2 NOx removal efficiency

The results of the photocatalytic efficiencies of the developed concrete under ISO conditions are presented in Figure 4.

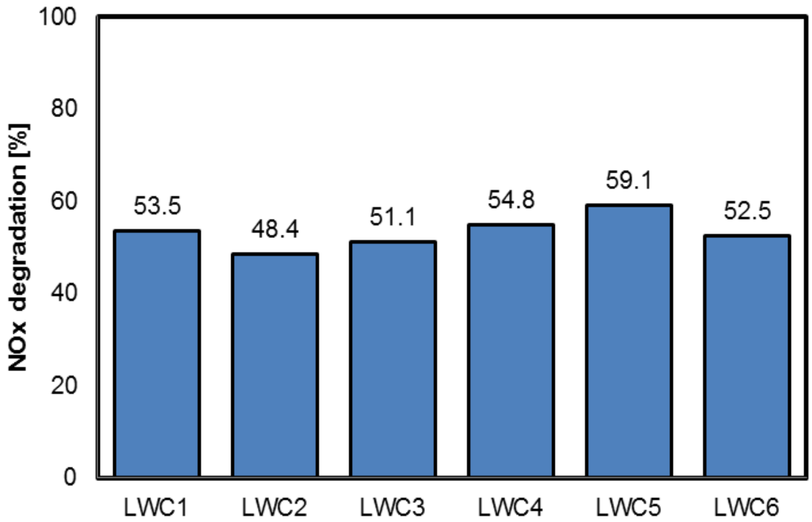

Figure 4: NOx removal efficiency results under ISO conditions.

The results show very high NOx degradation efficiencies (ranging from 48.4% to 59.1%) in all the developed samples, indicating their excellent air purifying performance. Furthermore, the samples containing zeolite (Mix 4-6) show in average an increase of 8% in de-NOx efficiency.

The results of the photocatalytic efficiencies under lower flow rate of 1.5 L/min but with same concentration (1.0 ppm) (UV1 in Table 2) conditions are presented in the Figure 5.

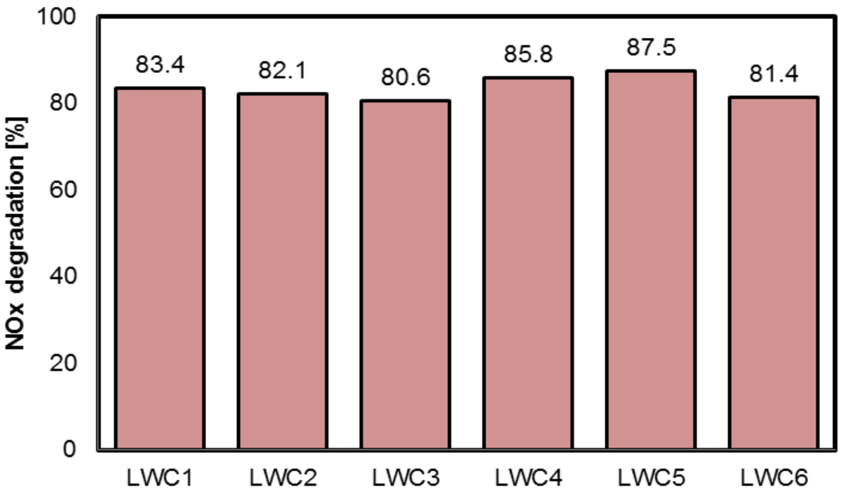

Figure 5: NOx removal efficiency results under UV1 conditions.

The lower flow rate promoted the efficiency significantly by increasing the residence time of the pollutant from 1.2 to 2.4 seconds. This resulted in excellent NOx removal efficiencies of over 80% in all cases.

The addition of zeolite promoted the photocatalytic efficiency (Mix 4-6) in case of all designs. The addition of lightweight aggregates led to decrease of efficiency in case of the recipes without zeolite. However, low amount of the light-weight aggregates (i.e. Mix 5) promoted the photocatalytic efficiency in case of recipe containing zeolite, leading to the highest NO_x degradation of 59.1% under the flow rate of 3L/min and 87.5% under the flow rate of 1.5L/min. This is attributed to the enhanced absorption ability by the applied zeolite together with the increased meso-porosities of the developed concrete.

Furthermore, Mix 5 is subjected to experiments under various environmental conditions, as presented in Table 2. The summary of the NO_x degradation efficiencies of the Mix 5 tested under ISO, UV1, UV2, UV3 and VIS1 conditions is presented in Figure 6.

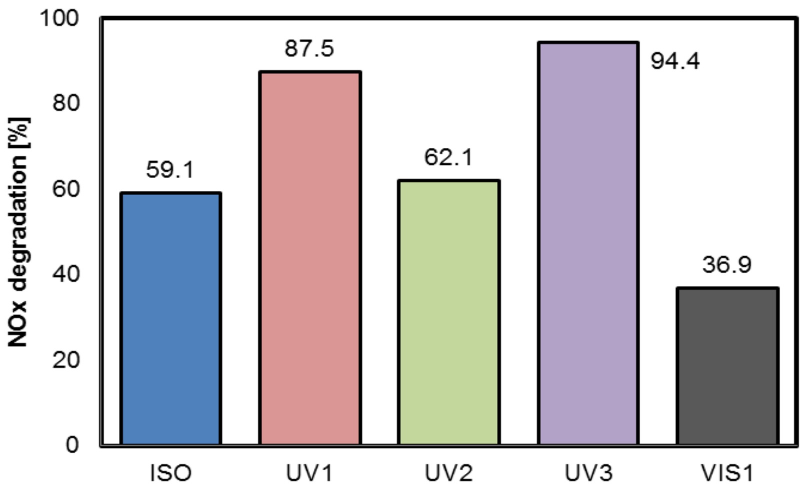

Figure 6: NOx removal efficiency of Mix 5 under various conditions.

The applied experimental conditions proved to be very crucial in affecting the photocatalytic efficiency. The applied lower flow rate of 1.5 L/min that represents closely the outdoor conditions resulted in significantly higher degradation efficiencies due to the increased residence time of the pollutant. However, the high concentration of 1.0 ppm did not represent the real outdoor/indoor conditions. When a lower concentration of 500 ppb was applied, almost all of the pollutant was degraded by Mix 5 (NO_x degradation of 94.4% under a flow rate of 1.5 L/min), which confirms again the very high efficiency of the developed material. Furthermore, the efficiency under indoor air conditions, i.e. under visible light irradiation was found to be very good as well (NOx removal of 36.9% under a flow rate of 1.5 L/min and an initial concentration of 500 ppb). This indicates a very promising potential of applying this product for indoor air quality improvement.

The tank reactor employed in this study is to further simulate indoor environment. An air exchange rate of 2 h^{-1} under the used air flow rate condition (1.5 L/min) is resulted and this represents a more realistic indoor air condition. In addition, the reactor volume to active surface ratio (4 dm^2 : 45 dm^3) represents much better realistic conditions here. The volume of the air is allowed to flow, not only above the surface of the sample, but also around it. In order to simulate the movement inside the reactor, the tank reactor is equipped with a ventilator. This ventilator is fully controllable. The inlet in the tank reactor is expanded, comparing to the plug-flow reactor, in order to ensure better air dispersion. Due to the higher volume, the air exchange in the reactor can be set to values closely representing indoor conditions.

Figure 7 shows the test results using the tank reactor condition under visible light irradiation. The results show lower degradation efficiency (25.7% compared to 36.9% in the case of Mix 5), which was expected due to much smaller active surface to volume ratio (2 dm^2 : 0.06 dm^3 in the case of ISO reactor condition). Nevertheless, it can be seen that under this condition the developed concretes show promising air pollutants removal ability under indoor visible light condition.

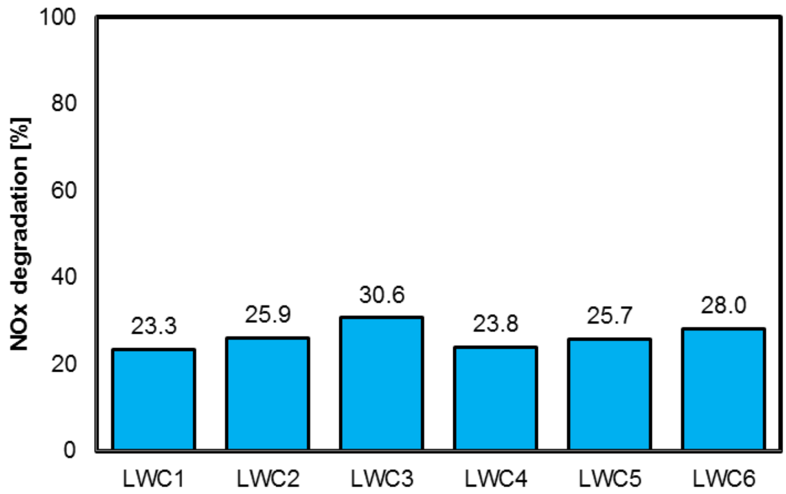

Figure 7: NO_x removal in the tank reactor under visible light irradiation.

4. Conclusions

This study aimed to develop a functional lightweight concrete with air purifying properties. Heterogeneous photocatalysis technology is applied in this development. Several recipes were designed, casted and tested for air pollutants removal efficiency by means of nitric oxide degradation following the ISO 22197-1 as a reference. The effect of porous materials such as lightweight aggregates and an absorptive material zeolite, on the photocatalytic efficiency was studied. Furthermore, a tank reactor with a scaled volume of 45 l is applied to better simulate an indoor environment to further evaluate the performance of the developed concrete. The following conclusions are reached:

- All the developed samples show excellent NO_x removal efficiency tested under ISO standard conditions (over 50% degradation in overall);
- Zeolite addition shows a positive effect on the photocatalytic efficiency leading to enhanced NO_x removal efficiencies;
- Samples containing a combined low amount of lightweight aggregates together with zeolite leads to the highest degradation efficiency;

- The developed samples demonstrate excellent air pollutants removal efficiency under realistic outdoor conditions (e.g. 94.4% NOx removal by Mix 5)
- The developed concrete also possesses very good air purifying performance under realistic indoor conditions (e.g. 37% NOx removal by Mix 5).
- All developed samples showed very good NOx removal efficiency using tank reactor under visible light conditions (around 25%)
- Tank reactor tests seem to be more suitable way of testing passive photocatalytic materials for real application compared to the ISO 22197-1 method.

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