van Lieshout, B.; Spiesz, P.; Brouwers, H.J.H.

Application of waste glass in translucent and photocatalytic concrete

Abstract

Container glass aggregates and glass powder are waste products of the glass recycling industry. In this research, these products are incorporated in self-compacting concrete (SCC) mixtures, replacing conventional aggregates and fine powders. The SCC mixtures were designed using a particle packing optimization algorithm in order to obtain good properties in both fresh and hardened states. Different mixtures were tested for their strength, alkali-silica reaction (ASR), translucency to the visible light and photocatalytic oxidation (PCO) properties. Results show that the strength of the SCC mixtures containing the waste glass, compared to a reference mixture prepared with conventional aggregates and sand, is slightly lower in all the investigated mixtures. With respect to ASR test results, mixtures with glass aggregates can be classified as highly reactive, but the glass powder can suppress the ASR. Transparency of the prepared concrete plates with glass aggregates is low. The addition of a photocatalyst was studied, to render the material self-cleaning and to promote photocatalytic oxidation (PCO) of NOx. The PCO test results show an improvement of the NOx degradation up to 40% compared to the reference samples without glass particles.

Introduction

Concrete is one of the most used materials in construction. The main reasons for that are its low price, high mechanical strength, high durability and variety of its form due to on-site casting or prefab design. Unfortunately these come at the cost of the application of large quantities of unsustainable elements like cement and aggregates. Extensive research has already been done on the replacement of cement with more sustainable materials. The aggregates consist of coarse and fine gravel and sand. These elements are unsustainable because in order to obtain them, often large areas of quarries are needed. To make the concrete mixture more sustainable, aggregates can be replaced (partially or completely) by container waste glass. Unfortunately in this case some harmful reactions can occur, such as alkali-silica reactions (ASR) in which a formed gel expands over a longer period of time, damaging the concrete. Even though extensive research has already been done on the alkali-silica reaction issue in concrete, it is still not completely clear how this reaction takes place. Most of the proposed ASR mechanisms are based on the same principle [1-4]: a reaction between the alkalis (Na2O and K2O) originating from the cement with silica originating from aggregates. As the product of this reaction, a gel of alkali-silicate is formed. This gel binds water and can expand to about twice its initial volume. This reaction only takes place if the amount of Na2O and K2O is high enough and when water is present [1]. Knowing how reactive different mixtures and materials are is very important because the expansion can deteriorate concrete in the
long term. Therefore, the application of low-alkali cements and/or non-reactive aggregates is crucial from the durability point of view. Different properties of materials used in concrete have an effect on the ASR reactivity. In the case of glass this comes to its particle size and chemical composition. For container waste glass, differences in ASR reactivity can be ascribed to different colors of the glass, i.e. different chemical compositions of the glass [5]. Besides the composition, also the particle size plays a role in the reactivity of the glass. It is found that larger particles have the long-term reactivity (deterioration capacity) higher than smaller particles [6]. When particles smaller than sieve mesh #50 (ASTM) are used, the produced mixtures have very low reactivity [7]. The reason for this difference in reactivity is the so called “pessimum effect” [1]. In general, the ASR is a very slow reaction (it can take years). When reactive fine materials (e.g. glass powder) are added, a large reactive surface is available. The larger the area, the faster the reaction takes place and completes (particles smaller than 1 mm are recommended [8]). If enough fine material (sufficient surface area) is available, the ASR can be finished before the hardening of concrete finishes. This means that the long-term expansion due to the ASR can be prevented. Because of the TiO₂-based photocatalytic oxidation (PCO) activated with the UV light, concrete may become a self-cleaning material. Additionally, if the right type of TiO₂ is applied, concrete can be used as an active air purifying material in both outdoor (activated by the UV-light) and indoor (activated by the visible light) applications [9]. Due to the applied glass, it might be possible that even more UV light is transferred to the TiO₂ particles, making their activation more efficient [10] and therefore this effect is also investigated in this study.

Materials and mix design

Materials

The glass used in this research project originates from a glass sorting facility (Maltha-groep B.V. the Netherlands). All of these samples are waste products of this facility which could not be further used for recycling. Two different forms of glass are used, coarse aggregates and powder. Two different types of coarse glass aggregates used were: a) color: mixed glass consisting green, brown, clear and blue glass particles and b) clear: glass consisting of only clear aggregates. To remove the organic residue both glass types were washed with water. Also a sample with unwashed glass was prepared in this study for investigating its influence on concrete properties.

<table>
<thead>
<tr>
<th>Oxide</th>
<th>Color [%]</th>
<th>Clear [%]</th>
<th>Powder [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>72.9</td>
<td>73.5</td>
<td>63.9</td>
</tr>
<tr>
<td>Na₂O</td>
<td>12.5</td>
<td>12.3</td>
<td>10.2</td>
</tr>
<tr>
<td>CaO</td>
<td>10.5</td>
<td>10.6</td>
<td>10.5</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>1.6</td>
<td>1.4</td>
<td>2.1</td>
</tr>
<tr>
<td>MgO</td>
<td>1.2</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>Cr₂O₃</td>
<td>0.1</td>
<td>0.009</td>
<td>0.1</td>
</tr>
</tbody>
</table>

Figure 1.
Ternary phase diagram of binders and by-products
The chemical composition of the glass is shown in Table 1. Based on this composition, Figure 1 shows where the used glass is located in a ternary phase diagram. River aggregates with sizes between 2-8 mm are used as reference non-reactive aggregates. This material covers about the same particle size range as the used glass aggregates, as shown in Figure 2. Microsand (Granite Import Benelux, the Netherlands) is used as an inert filler. Microsilica is used to investigate its effect on the ASR suppression. This silica is applied as 50% solid suspension in water. For aesthetic reasons of the final product, white Portland limestone cement CEM II/A-LL 42.5N is used in this study. This cement consists of 80-94 % OPC clinker, 6-20 % limestone powder and has a content of alkalis lower than 0.60%. The particle size distributions (PSDs) of all used solid materials are shown in Figure 2.

**Figure 2.**
Particle size distribution of used solid materials

**Mix design**
The SCC mixes were designed using a particle packing optimization algorithm described in [11], which employs the modified Andreasen and Andersen particle packing model, as shown in the following equation:

$$ P(D) = \frac{D^q - D_{\text{min}}^q}{D_{\text{max}}^q - D_{\text{min}}^q} \quad \forall D \in [D_{\text{min}}, D_{\text{max}}] $$  \hspace{1cm} (1)

where: \( D \) – particle size [μm], \( P(D) \) - cumulative passing fraction through a sieve with the opening size \( D \), \( q \) – distribution modulus, \( D_{\text{min}} \) – minimum particle size [μm] and \( D_{\text{max}} \) – maximum particle size [μm].

The proportions of ingredients used to produce the designed SCC mixtures are presented in Table 2, in which the SP corresponds to the superplasticizer amount and w/p to the water/powder ratio (powder is defined as particles smaller than 250 μm). The SP amount and the water contents in the different SCC mixtures were varied in order to maintain the same flowability spread (about 35 mm using the Hägerman cone).
Table 2.
Mix proportions of SCC (kg/m³)

<table>
<thead>
<tr>
<th>Mix</th>
<th>CEM II/A-LL</th>
<th>Microsilica</th>
<th>Microsand</th>
<th>Glass powder</th>
<th>Coarse aggregates</th>
<th>Type of coarse aggregates</th>
<th>Washed</th>
<th>SP [wt % of cement]</th>
<th>w/p</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>400</td>
<td>-</td>
<td>571.6</td>
<td>-</td>
<td>845.1</td>
<td>Clear glass</td>
<td>Yes</td>
<td>0.32</td>
<td>0.32</td>
</tr>
<tr>
<td>B</td>
<td>400</td>
<td>-</td>
<td>585.5</td>
<td>-</td>
<td>807.7</td>
<td>Color glass</td>
<td>No</td>
<td>0.39</td>
<td>0.33</td>
</tr>
<tr>
<td>C</td>
<td>400</td>
<td>-</td>
<td>585.5</td>
<td>40.9</td>
<td>809.8</td>
<td>Color glass</td>
<td>Yes</td>
<td>0.45</td>
<td>0.29</td>
</tr>
<tr>
<td>D</td>
<td>350</td>
<td>-</td>
<td>589.4</td>
<td>-</td>
<td>804.2</td>
<td>Color glass</td>
<td>Yes</td>
<td>0.28</td>
<td>0.35</td>
</tr>
<tr>
<td>E</td>
<td>400</td>
<td>32.5</td>
<td>547.9</td>
<td>-</td>
<td>793.9</td>
<td>Gravel</td>
<td>-</td>
<td>0.29</td>
<td>0.30</td>
</tr>
<tr>
<td>F</td>
<td>400</td>
<td>-</td>
<td>630.9</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.16</td>
<td>0.31</td>
</tr>
</tbody>
</table>

Test Methods

Strength
Strength measurements were performed at 1, 7 and 28 days after casting. For each mixture three prisms (40 × 40 × 160 mm) were used. After casting, the molds were covered with plastic in order to prevent the evaporation of water. One day after, the samples were demolded and subsequently stored in water at room temperature. The three point flexural bending strength test was performed using a computer controlled testing machine. After this test, both halves of the broken prisms were used for the compression strength test.

Alkali-silica Reaction
For each mix three prisms (40 × 40 × 160 mm) with embedded stainless steel pins were cast and used for the ASR test, following the description given in [12]. Each prism was uprightly placed in a plastic container with a volume of approximately 1.6 liter. First the prisms were placed in a container with water and stored in an oven at 80°C. After 24 hours, the length of each prism was measured (zero reading between the embedded steel pins), and then the samples were transferred to a container with a 1M NaOH solution and stored in an oven at 80°C. The prisms stayed in these conditions for 14 days, during which their lengths were measured periodically. Figure 3 shows the ASR test set-up.

Figure 3.
Expansion measurement test set-up, storage container and oven for the ASR test
Photocatalytic Oxidation

The experimental conditions applied in this study (shown in Table 3) were in accordance with [13]. A detailed description of the used test set-up and test procedure is given in [9]. Two different plates (200 × 100 × 30 mm) were cast for the PCO: a reference sample with gravel (Mix F in Table 2) and a sample with glass aggregates (Mix C in Table 2). In both samples 5% TiO₂ by weight of cement was applied. After the casting and curing period (28 days), both samples were tested from two sides: top surface in its original cast form and the bottom mold-surface after an additional polishing. The unpolished sides have a rough surface, so there was a larger exposed surface area of the cement paste. The polished sides have the cement paste layer removed, exposing the aggregates.

Table 3.
Photocatalytic oxidation test conditions

<table>
<thead>
<tr>
<th>NO concentration</th>
<th>1000 ppb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Transport medium</td>
<td>Synthetic air</td>
</tr>
<tr>
<td>Reactor’s height</td>
<td>3 mm</td>
</tr>
<tr>
<td>Exposed surface area of the sample</td>
<td>200 × 100 mm</td>
</tr>
<tr>
<td>Flow rate</td>
<td>3.0 [l/min]</td>
</tr>
<tr>
<td>Humidity</td>
<td>50 % RH</td>
</tr>
<tr>
<td>UV-Irradiance</td>
<td>10 [W/m²]</td>
</tr>
</tbody>
</table>

Translucency
Concrete plates composed of only glass aggregates and cement paste were cast. After the curing period they were polished to expose the glass aggregates and remove the cement paste from the surface layers. After this first rough-polishing step, the plates have been subjected to extra fine-polishing steps. In this way their aesthetic appearance was improved. The light transmittance through the plates was examined by comparing images taken with a photo camera.

Results and discussion

Strength
As can be seen in Figures 4 and 5, the average compressive and flexural strengths of the prepared SCC concrete samples containing glass aggregates were slightly lower compared to the reference samples without the glass. This can be attributed to a lower bonding strength between the aggregates and the cement matrix and to the lower strength of the glass itself compared to conventional gravels and sands. The mixture containing the glass powder (mix D) has a substantially lower strength compared the other mixtures. This could be attributed to the lower cement amount used in that mixture and to the pollution of the glass powder with organic matter. As expected, the mixture with microsilica (Mix E) reached the highest strength among the samples containing the glass aggregates due to additional cementitious properties of the microsilica. Also a difference between the washed and unwashed glass particles used in the mixtures is found. The mixture containing the washed glass aggregates has a slightly higher strength because the organic matter was removed.
Alkali-silica reaction

The ASR test results of the concrete prisms are presented in Figure 6. According to [14], the total expansion of less than 0.1% (160 µm for 160 mm initial length of the prisms) at the age of 14 days means that the tested sample is not very reactive. As expected, the reference mixture (without glass) shows the lowest expansion, fulfilling the 0.1% expansion criterion. However, Mix B in which the glass powder was applied together with the coarse glass aggregates, shows an expansion similar to the reference mixture. This reflects on a suppression of the ASR by application of the fine glass powder. On the other hand, the microsilica applied in Mix E did not help to reduce the ASR. All other tested samples show a high ASR reactivity, failing the 0.1% maximum expansion criterion.

Photocatalytic Oxidation (PCO)

The PCO efficiency of NOx (NO and NO2) removal from the air is shown in Table 4. Although the same amount of TiO2 was applied in both mixtures (TiO2-Gravel being the
reference mixture where a conventional gravel and sand were used and TiO$_2$-Glass where the glass aggregates were used), the plates containing glass aggregates show higher pollutants degradation efficiency ($D_{NOx}$) compared to the plates without these aggregates. Regardless of the type of exposed surface (rough, covered with cement paste and having a larger specific surface area and polished, with a lower surface area and exposed aggregates), the reference plates with the gravel aggregates show very comparable pollutants decomposition efficiencies. This is unexpected because there was less exposed TiO$_2$ at the polished surface compared to the rough surface, and therefore less TiO$_2$ particles could be activated with the UV light. The samples containing the glass aggregates show significantly improved PCO efficiency (about 40%) compared to the reference samples without the glass. Additionally, for the plate containing glass aggregates the PCO efficiency differed between the polished and unpolished surfaces: the unpolished plate performs about 20% better. This can be explained by the fact that the unpolished sample benefits from both: a) more surface area compared to the polished plate and b) the glass particles underneath the surface (cement paste layer) could still transfer the UV light to activate the TiO$_2$. The unpolished plate with glass aggregates performs the best in the test series, with the $D_{NOx}$ rate of 52.53%.

Table 4. Photocatalytic oxidation test results

<table>
<thead>
<tr>
<th>Sample description</th>
<th>TiO$_2$-Gravel</th>
<th>TiO$_2$-Gravel</th>
<th>TiO$_2$-Glass</th>
<th>TiO$_2$-Glass</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polished</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>$D_{NOx}$ [%]</td>
<td>37.61</td>
<td>37.42</td>
<td>43.18</td>
<td>52.53</td>
</tr>
</tbody>
</table>

Translucency

The translucency of the produced plates can be an attractive added feature from the point of view of aesthetical and architectural applications. Figure 7 and 8 show that, for similar mixtures, the resulting visible light translucency may be very different. This can be attributed to the casting technique, as the best translucency can be obtained when the elongated glass particles are positioned perpendicularly to the larger surfaces of the plates. The plates produced in this study have relatively low mechanical properties (especially against bending and impact) due to their small thicknesses (about 10 mm). These thicknesses were limited by the maximum grain size of the used glass aggregates, because in order to obtain a translucent spot within the plate, the same glass aggregate should be exposed from both sides of the plate. In order to obtain thicker (stronger) samples, larger fractions of the glass aggregates should be used.
Conclusions

Glass aggregates can be used as replacement of conventional aggregates in concrete. Sufficient mechanical properties of the produced SCC containing glass aggregates were obtained in this study, using economical amounts of cement. However, the alkali-silica reaction may be a practical durability issue when applying glass aggregates in concrete. On the other hand, it is shown in this study that the fine glass powder may be successfully used to suppress ASR. Translucency of the product can be an attractive feature for concrete containing glass aggregates. However, for small-sized glass aggregates, the produced plates may be quite fragile, but this drawback can be improved using larger size fractions. Additionally, the application of the glass aggregates significantly improves the efficiency of the PCO of pollutants, due to a higher energy being transferred through the glass to activate TiO$_2$ particles.

References

[1] Vegt, I. (2003), Graduation report: Glas in beton. Delft University, the Netherlands
[4] Bulteel, D. et al. (2000), Alkali-aggregate reaction: a method to quantify the reaction degree. Proceedings 11th International Conference on Alkali Aggregate Reaction, Quebec, Canada

Authors

MSc. Bart van Lieshout
MSc. –Eng. Przemek Spiesz
Prof.dr.ir Jos Brouwers
Eindhoven University of Technology
Faculty of the Built Environment, P.O. Box 513
5600 MB Eindhoven
The Netherlands
E-mail: p.spiesz@tue.nl