Alkali activated ultra-lightweight concrete: an innovation on functionality and sustainability

Citation for published version (APA):

Document status and date:
Published: 01/01/2015

Document Version:
Accepted manuscript including changes made at the peer-review stage

Please check the document version of this publication:

• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
• The final author version and the galley proof are versions of the publication after peer review.
• The final published version features the final layout of the paper including the volume, issue and page numbers.

Link to publication

General rights:
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
• You may not further distribute the material or use it for any profit-making activity or commercial gain
• You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the “Taverne” license above, please follow below link for the End User Agreement:
www.tue.nl/taverne

Take down policy:
If you believe that this document breaches copyright please contact us at:
openaccess@tue.nl
providing details and we will investigate your claim.
Alkali activated ultra-lightweight concrete: an innovation on functionality and sustainability

1. Introduction

Alkali activated materials (AAM), in many cases interpreted using another term geopolymer, have attracted more and more attention due to the greatly reduced environmental impact they contribute compared to the ordinary Portland cement. This type of material generally exhibits better performances such as mechanical properties, durability, thermal properties and lower environmental impacts when compared to ordinary Portland cement. They can be classified into two types according to the calcium content in the raw materials. One is the high calcium system, having a C-A-S-H type gel as the main reaction product [1]. The other is the low calcium system, having N-A-S-H type gels with three-dimensional network as the major reaction product [2]. Recently attention is paid to blended alkaline systems that are produced by mixing calcium enriched precursors with aluminosilicates due to several modified properties such as setting times, workability, shrinkage, mechanical properties and durability [3], and promising progresses in understanding the blended system and those modified properties indicate a promising future [4;5]. Furthermore, the concept of ultra-lightweight concrete has great potential because of the flexibility it brings in terms of architectural design, mechanical property, thermal insulating property and durability. In the previous research, a mix design methodology was proposed to design ultra-lightweight concrete and results indicate the excellent performance of the developed concrete [6].

This research addresses the development of an alkali activated ultra-lightweight concrete (termed Geo-ULC), aiming at the application in monolithic concrete façade structure as both load bearing element and thermal insulator. The effect of the alkali activators type, including sodium hydroxide modified sodium silicate and sodium hydroxide modified sodium silicate further modified by sodium carbonate, on the properties of the developed concrete is investigated. The reaction kinetics of the alkali activation is evaluated by using isothermal calorimetry. Ground granulated blast furnace slag (GGBS) is used as the raw materials and moreover, substituting the GGBS by fly ash and limestone powder is researched.

2. Experiment

2.1 Materials

The solid materials used here are ground granulated blast furnace slag, class F fly ash and limestone powder. Their chemical compositions are shown in Table 1. The specific densities of GGBS, fly ash and limestone powder, determined using a gas Pycnometer (AccuPyc II 1340 Pycnometer), are 2924.6, 2301.5, and 2710 kg/m³, respectively. The median particle size (d50) is 12.43 μm for slag, 22.06 μm for fly ash and 10.12 μm for limestone powder.
Table 1: Major chemical compositions of raw materials

<table>
<thead>
<tr>
<th>Oxides (wt.%)</th>
<th>Fly ash</th>
<th>Slag</th>
<th>Limestone</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO$_2$</td>
<td>54.6</td>
<td>34.4</td>
<td>0.84</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>24.4</td>
<td>13.3</td>
<td>0.24</td>
</tr>
<tr>
<td>CaO</td>
<td>4.44</td>
<td>37.4</td>
<td>53.96</td>
</tr>
<tr>
<td>MgO</td>
<td>1.43</td>
<td>9.89</td>
<td>1.01</td>
</tr>
<tr>
<td>Fe$_2$O$_3$</td>
<td>7.2</td>
<td>0.47</td>
<td>0.32</td>
</tr>
<tr>
<td>Na$_2$O</td>
<td>0.73</td>
<td>0.34</td>
<td>0.21</td>
</tr>
<tr>
<td>K$_2$O</td>
<td>1.75</td>
<td>0.47</td>
<td>0.34</td>
</tr>
<tr>
<td>SO$_3$</td>
<td>0.46</td>
<td>1.23</td>
<td>-</td>
</tr>
<tr>
<td>LOI</td>
<td>2.80</td>
<td>1.65</td>
<td>43.01</td>
</tr>
</tbody>
</table>

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, which provide a great freedom for the design of lightweight concrete with desired low density, as can be seen in Table 2.

![Figure 1: SEM pictures of (a) external fracture surface, (b) internal structure.](image)

Table 2: Properties of the lightweight aggregates.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Bulk density (kg/m$^3$)</th>
<th>Specific density (kg/m$^3$)</th>
<th>Crushing resistance (N/mm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LWA A</td>
<td>300</td>
<td>540</td>
<td>&gt;2.9</td>
</tr>
<tr>
<td>LWA B</td>
<td>250</td>
<td>450</td>
<td>&gt;2.6</td>
</tr>
<tr>
<td>LWA C</td>
<td>220</td>
<td>350</td>
<td>&gt;2.4</td>
</tr>
<tr>
<td>LWA D</td>
<td>190</td>
<td>310</td>
<td>&gt;2.2</td>
</tr>
<tr>
<td>LWA E</td>
<td>170</td>
<td>300</td>
<td>&gt;2.0</td>
</tr>
</tbody>
</table>
The activator used includes (a) sodium hydroxide pellets (analytical level) modified sodium silicate solution (27.69% SiO₂, 8.39% Na₂O and 63.92% H₂O by mass). (2) sodium hydroxide pellets (analytical level) modified sodium silicate solution (27.69% SiO₂, 8.39% Na₂O and 63.92% H₂O by mass), further modified by Na₂CO₃ (analytical level). The desired activator modulus (Ms, SiO₂/Na₂O molar ratio) was achieved by adding different amount of sodium hydroxide pellets into the sodium silicate solution. The required Na content is calculated from either the Na containing in (1) NaOH and Na₂SiO₃, (2) NaOH and Na₂SiO₃ and Na₂CO₃. The mixed activator solution was cooled down to room temperature prior to further use. Distilled water was added in order to reach the desired water/binder ratios. An air-entraining agent (Cugla), with a density of 1.05 kg/l and resin acid soaps as active agent, is applied here to adjust the density of the lightweight concrete.

2.2 Mix design

A mix design methodology previously used for normal density mortars and concretes was considered for the design of the alkali activated ultra-lightweight concrete. This mix design tool is based on the insight that superior properties of a granular mix are achieved when a so-called geometric grading curve is designed and obtained. In the case of continuous distributions, the cumulative finer fraction of the entire mix is determined from the design method proposed by [7], reads:

\[ P(D) = \frac{D^q - D^q_{\text{min}}}{D^q_{\text{max}} - D^q_{\text{min}}} \]  

(1)

where \( P(D) \) is a fraction of the particles being smaller than size D, D is the particle size (μm), \( D_{\text{max}} \) and \( D_{\text{min}} \) are the largest and smallest particle size (μm), respectively, in the mix, and q is the distribution modulus.

This particle packing principle insight has been transformed into a numerical mix design, in which all the solid mixture ingredients, which all have their own particle size distributions (PSDs), are combined via a mathematical optimization routine, i.e. the “target curve” is approached best. The optimization of the particle size grading of the ingredients helps to increase the packing of the solids in the concrete mixture. This results in improved hardened state properties, a reduced water demand as well as an improved workability, since more water is available to act as lubricant between the particles [8]. This design methodology has been successfully applied to design earth-moist concrete [9], self-compacting concrete [10], gypsum based composite [11], lightweight concrete [12] and high performance concrete [13].

In addition to the targeted strength, a low thermal conductivity is also aimed in this study, while it is strongly related to the density and porosity of the composite [14]. Hence, a densified concrete developed applying this design concept will show an opposite effect, i.e. the resulting thermal conductivity will increase as the void content is minimized. Nevertheless, the low density will be achieved by applying a lightweight material as aggregate (LWA) instead of normal density aggregates, and furthermore by applying an air-entraining agent. Three recipes were designed and the alkali modulus, Na content and water dosage were determined based on the preliminary research, as listed in Table 3.
Table 3: Mix design of the ULWC (kg/m³).

<table>
<thead>
<tr>
<th>Materials</th>
<th>Mix 1</th>
<th>Mix 2</th>
<th>Mix 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>GGBS</td>
<td>243.38</td>
<td>466.5</td>
<td>233.25</td>
</tr>
<tr>
<td>Fly ash</td>
<td>146.02</td>
<td>0</td>
<td>93.3</td>
</tr>
<tr>
<td>Limestone powder</td>
<td>97.34</td>
<td>0</td>
<td>139.95</td>
</tr>
<tr>
<td>Waterglass</td>
<td>84.38</td>
<td>96.04</td>
<td>96.04</td>
</tr>
<tr>
<td>NaOH</td>
<td>15.98</td>
<td>7.66</td>
<td>7.66</td>
</tr>
<tr>
<td>Na₂CO₃</td>
<td>0</td>
<td>23.92</td>
<td>23.92</td>
</tr>
<tr>
<td>Water</td>
<td>143.26</td>
<td>151.6</td>
<td>151.6</td>
</tr>
<tr>
<td>LWA A</td>
<td>26.36</td>
<td>26.36</td>
<td>26.36</td>
</tr>
<tr>
<td>LWA B</td>
<td>39.54</td>
<td>39.54</td>
<td>39.54</td>
</tr>
<tr>
<td>LWA C</td>
<td>30.34</td>
<td>30.34</td>
<td>30.34</td>
</tr>
<tr>
<td>LWA D</td>
<td>53.32</td>
<td>52.32</td>
<td>52.32</td>
</tr>
<tr>
<td>LWA E</td>
<td>63.7</td>
<td>63.7</td>
<td>63.7</td>
</tr>
<tr>
<td>Air entraining agent</td>
<td>0.657</td>
<td>0.675</td>
<td>0.675</td>
</tr>
</tbody>
</table>

2.3 Experimental program

The reaction kinetics was studied by an isothermal calorimeter. The solid raw materials were firstly mixed with the activating solution externally for about 1 min with vibration, then the mixed paste was injected into an ampoule and loaded into the calorimeter. All measurements were conducted for 120 h under a constant temperature of 20 °C. Further experiments were performed on both fresh and hardened concrete. The slump tests of the fresh concrete were carried out following EN 12350-2:2009 [15]. The flow table tests were performed following EN 12350-5:2009 [16]. The density of the fresh concrete was determined following EN 12350-6:2009 [17], using a container with a volume of 8.0 dm³. The air content in fresh state was determined experimentally. Firstly the fresh mixes were poured into a cylindrical container of a known volume, followed by compaction on a vibration table. Then, the mass of the concrete was measured and, assuming that the fresh concrete is homogeneous, the air content was derived from the density difference between the designed and prepared concrete. Samples with the size of 150 × 150 × 150 mm³ were then cast for further testing. The LWAC was produced using a planetary concrete mixer following a normal concrete production procedure 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°C), following EN 12390-2:2000 [18], until the test age was reached.

The apparent density of the samples in hardened state was determined in both ambient conditions and oven dry conditions, by calculating from the measured size and mass of the samples. The compressive strength tests were performed on cubes with the size of 150 × 150 × 150 mm³ by applying a load rate of 10.0 kN/s until fracture. Prior to the thermal conductivity measurement, the samples were dried in a ventilated oven at 105 °C until a constant mass, following EN 12390-7:2009 [19]. Then, the samples were cooled down to the room temperature. Subsequently, the thermal conductivity tests were performed on these samples employing a heat analyzer. The analyzer applies a dynamic measurement method to determine simultaneously the volumetric heat...
capacity \((J/(m^3\cdot K))\) and the thermal conductivity \((W/(m\cdot K))\) of materials with a measurement time of about 8-16 minutes. The measurement is based on the analysis of the temperature response of the tested sample to heat flow impulses, while the heat flow is excited by electrical heating of a resistor heater inserted into the probe which is in direct contact with the test sample.

3. Results and discussion

3.1 Reaction kinetics

The normalized heat flows of the new binders within the test period are shown in Figure 2.

![Figure 2: Normalized heat flow of the applied alkali activated binders (a) blended raw materials activated by water glass, (b) GGBS activated by ternary activator.](image)

The presented calorimetric curves are in accordance with the previous studies on silicate activated slag or its mixtures [5], which show four typical reaction stages including initial dissolution, induction, acceleration and stable period. The acceleration peaks of the two mixtures are located at different times of about 30 and 14 hours after mixing, respectively. This is assigned to the massive formation of reaction products from dissolved Ca, Si and Al units. It is clearly shown that when the raw materials are blended GGBS, fly ash and limestone powder, the heat evolution peak significantly shifts to later locations, surprisingly both show similar intensities. This demonstrates
the remarkable effects of the slag content on the early age reaction. Under alkali activation, the breakdown of a calcium enriched structure is easier than Si and Al dominated ones such as fly ash. Thus a higher slag content will lead to a larger amount of available Si, Al and Ca units in solution, and consequently a more intense dissolution and faster reaction process.

3.2 Fresh state behaviour

In overall, all the developed mixes show very good workability, no traces of segregation or bleeding were observed while performing the workability tests. The slump results show that Mix 1 (110 mm) can be classified in the slump class of S3 and Mix 2 (270mm) in S5, according to EN 206-1:2000. The flow table test results show that Mix 1 (550 mm) falls under the flow class of F4 and Mix 2 (640 mm) under F6. Here especially Mix 3 shows an excellent workability, with a flow of 670 mm without jointing that shows self-compacting characteristics. The better workability in the case of Mix 3 can be explained by the incorporated fly ash and limestone powder, as confirmed in a previous research [5].

The fresh state densities of the concrete mixtures are calculated from the measured mass and the fixed volume, yielding between 950 kg/m$^3$ (Mix 1), 890 kg/m$^3$ (Mix 2) and 830 kg/m$^3$ (Mix 3), respectively. The determined air contents in the developed ULWAC show 0 (Mix 1), 6.7% (Mix2 ) and 11.9% (Mix 3), respectively. This indicates the used Na$_2$CO$_3$ may have an positive effect on the efficiency of the air entraining agent, this might be explained by the reduced pH condition in the prepared alkali activator, which is also confirmed by Mix 1 as under a high pH condition the air entraining agent is not effective. The air content is higher in Mix 2 compared to that of Mix 3, which can be explained by the better workability of Mix 3 that contributes to a higher efficiency of the used air entraining agent.

![Figure 3](image)

Figure 3: Cross-section of the developed concrete.

3.3 Microstructure

As already explained, in the development of lightweight concrete, a proper spatial distribution of lightweight aggregates is crucial especially when LWAs of a very low density are used. In the present study the used LWAs have a very low density, as listed in Table 2, holding in the range of 300 - 540 kg/m$^3$, indicating a very high segregation
potential of the cement paste from the LWA if the mixture is not properly designed. Therefore, the distribution of the LWAs in the concrete matrix was investigated. Figure 3 shows the cut surface of the selected samples after performing the compressive strength test. As can be seen in this figure, in all the developed mixtures the LWAs are very homogeneously and evenly distributed in the concrete matrix. This confirms that there was no segregation in the mixtures developed in the present study, indicating the suitability of the applied concrete design method as well as the sufficiently high viscosity of the binder paste.

3.4 Compressive strength

The 7 and 28 day compressive strength of the developed ultra-lightweight concrete are shown in Figure 4. All the three concretes show a compressive strength of over 10 N/mm² at 28 days. In addition, as can be seen in Figure 4, the compressive strength of all the samples at the age of 7 days has already reached over 85% of their strengths at 28 days. This is in line with the authors’ previous study [19] and also with [1] where cement is applied as binder. Mix 1 shows a slightly higher 28-day compressive strength compared to Mix 2 and Mix 3. This can be explained from the binder composition as in Mix 2 and Mix 3 Na₂CO₃ is applied as part of the alkali activator while from literature in general water glass provides the best activation efficiency in terms of strength. Under the same alkali activator, Mix 2 shows a faster strength development at the early age. This also confirms the low reactivity of fly ash, or the dissolved Si and Al units from fly ash exhibits much less influence than the Ca from slag on strength at the early age under ambient curing condition.

3.5 Thermal conductivity

All the three developed concrete shows very good thermal physical property, reflected by the very low thermal conductivities. The thermal conductivities of the three mixtures are 0.14, 0.13 and 0.13 W/(m·K), respectively, measured to the samples under oven dry conditions at 28 days age. Figure 5 provides a comparison between the compressive strength and thermal conductivities of different types of concrete available in the literature [12:20-23]. In this Figure, also the results of the previously developed ultra-lightweight concrete based on cement is shown. It can be clearly observed that the presented results in this study show excellent thermal properties, indicated by at this thermal conductivity range the compressive strength of the Geo-ULC is much
higher than other reported data. While it can also be seen that the results shown in this study fall into the same range of the results obtained from the ultra-lightweight concrete developed in the previous study, as shown in this Figure.

![Figure 5: Thermal conductivity vs. compressive strength](image)

### 4. Conclusions

This study presents the development of an alkali activated ultra-lightweight aggregates concrete (Geo-ULC), aiming at a good balance between mechanical properties and thermal conductivity, thus performing as both load bearing element and thermal insulator. Based on the presented study, the following conclusions can be reached:
- Alkali activated materials show their effectiveness as binder for developing ultra-lightweight concrete;
- The reaction kinetics of the designed binders are investigated and the influential factors are evaluated;
- The developed Geo-ULC has a very good workability and all the lightweight aggregates are homogeneously distributed in the concrete matrix;
- The developed Geo-ULC shows a 28-day compressive strength above 10 N/mm², and a thermal conductivity of about 0.13 W/(m·K).

### 5. Acknowledgements

The authors wish to express their gratitude to the following sponsors of the Building Materials research group at TU Eindhoven: Rijkswaterstaat Grote Projecten en Onderhoud; Graniet-Import Benelux; Kijlstra Betonmortel; Struyk Verwo; Attero; ENCI HeidelbergCement; Rijkswaterstaat Zee en Delta-District Noord; Van Gansewinkel Minerals; BTE; V.d. Bosch Beton; Selor; GMB; Geochem Research; Icopal; BN International; Eltoman;
Knauf Gips; Hess AAC Systems; Kronos; Joma; CRH Europe Sustainable Concrete Centre; Cement & Beton Centrum; Heros and Inashco (in chronological order of joining).
References


Authors:

Dr. Q.L. Yu MSc.; B. Yuan MSc.; X. Gao MSc.; Prof.dr.ir. H.J.H Brouwers
Department of the Built Environment
Eindhoven University of Technology
P.O. Box 513
5600 MB Eindhoven
The Netherlands
q.yu@bwk.tue.nl