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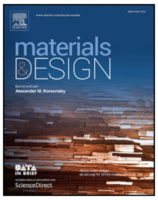

Design and performance evaluation of ultra-lightweight geopolymer concrete

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ABSTRACT

This article presents the development of a sustainable ultra-lightweight geopolymer concrete (with a dry density $\leq 800\ kg/m^3$) for both thermal insulating and load bearing purposes. A predesigned mineral precursor resulted from industrial by-products is used as the raw material for alkali activation and NaOH with a low concentration of 2–3 M is used as the alkali activator, together with a specially designed additive as superplasticizer. The ultra-lightweight property is achieved by applying a waste glass produced expanded lightweight material as aggregates. The effects of influential parameters including the design approach, liquid/binder ratio, binder/aggregate ratio, particle size and air entraining agent on the properties of the designed concrete are evaluated. An ultra-lightweight concrete with a proper workability is obtained and the hardened concrete shows excellence performances in terms of mechanical property, thermal property and durability. At a moderate compressive strength (10 MPa), an excellent thermal conductivity of 0.11 W/(m·K) is resulted. Furthermore, a very low thermal conductivity of 0.07 W/(m·K) is reached at a compressive strength of 8 MPa, indicating great potential for the production of a load bearing and highly insulating building material as an alternative for the traditional materials.

1. Introduction

Portland cement (PC) is the world's most used binder for production of construction materials. Due to its good mechanical property, relatively low cost, good durability and availability of the raw materials, PC concrete is favoured in many applications. Nevertheless, the production of PC has some major drawbacks, e.g. depletion of natural habitat and fossil fuels, and high emissions of CO_2 and other greenhouse gas [1]. Therefore, increasing interests on searching for alternative systems such as alkali-activated materials (AAM) (or in many cases interpreted as geopolymer) are observed [1]. These materials may be beneficial in terms of overall sustainability characteristics by applying industrial by-products as a partial precursor material instead of a primary raw mineral binder like PC. Additionally, depending on the applied raw materials and alkali activators, the produced end-products could show better properties compared to PC concrete, such as lower hydration heat, fast early strength development, stronger aggregate–matrix interface formation, lower thermal conductivity (TC) and higher acid and fire resistance [2–5]. Due to these unique material characteristics, increasing numbers of business startups of private and public institutes are seen, investigating and producing alkali activated materials.

In general two types of alkali activated materials can be classified: (a) the high calcium system, with ground granulated blast furnace slag (GGBS) as a typical precursor, having a C-A-S-H type gel as the main reaction product [6]; (b) the low calcium system with Class F fly ash and metakaolin as representative raw materials, having N-A-S-H type gels within a three-dimensional network as the major reaction product [7]. Extensive research has been performed on these two systems, including for instance the role of activator type and alkali concentration [8], the effect of the dosage of raw materials [9], the effect of admixtures [10–11], the curing effect [12], microstructure, mechanical properties, thermal properties and durability [13], etc. However, although excellent performances can be achieved from both systems, drawbacks such as fast setting, high shrinkage of alkali activated slag [14], elevated curing temperatures demand and relatively long setting times of alkali activated aluminosilicates limit their practical application. Research on overcoming these disadvantages is being widely explored and one promising solution is the blended alkaline systems ($Na_2O–CaO–Al_2O_3–SiO_2$ systems) that are produced by mixing calcium enriched precursors and aluminosilicates [15–20].

Lightweight aggregates concrete (LWAC) has its roots in the ancient period about 3000 years ago when volcanic materials were used as lightweight aggregates [21]. Because of its many advantages such as low density, good thermal insulation, good fire resistance and reduced cost of transport, LWAC has been widely investigated and developed in a wide range of unit weights and suitable strengths for various applications as both structural and non-structural material [22]. In the previous research, a mix design methodology based on the optimized packing principle was proposed for designing both structural concrete

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(with an oven dry density of about 1500 kg/m^3) [22] and non-structural concrete (with an oven dry density of about 650 kg/m^3, also called ultra-lightweight concrete) [23] using cement as the binder and the excellent tested results indicates the validity of the applied design approach as well as the superior performance of the developed concretes.

Applying alkali activated materials as the binder to develop lightweight concrete has been investigated. Recycled lightweight block was used as aggregates and alkali activators such as $Ca(OH)_2$, $Mg(NO_3)_2$, $Ca(OH)_2$ and Na_2SiO_3 and fly ash and GGBS as raw material in producing lightweight geopolymer concrete [24–25]. Results show at a density of about 1500 kg/m^3, a 28-day compressive of about 13 MPa is obtained, while at a density of about 1000 kg/m^3, a very low value of about 2 MPa is resulted [24]; and a 28-day compressive strength of about 0.5–2.0 MPa and thermal conductivity of about 0.09–0.13 W/(m·K) are achieved at a density range of 325–500 kg/m^3 [25]. Liu et al. [26] reported an average pore size of 1 mm and the compressive strength between 7 and about 10 MPa when the porosity decreases from 85% to 40% in a phosphoric acid based porous geopolymers using aluminium powder as a foaming agent. Lightweight concrete using activated metakaolin-fly ash as binder and aluminium powder to achieve the low densities was designed [27]. The compressive strength is strongly affected by the density, namely between 11.0 and 14.6 MPa at the density of 1200 kg/m^2 while 3.0–6.4 MPa at 900 kg/m^3 and only 1.3–1.8 MPa at 600 kg/m^3. Higher curing temperatures of up to 75 °C are observed to accelerate the early age strength development during the first day but in the long term no difference is observed. Thermal conductivity on the other hand, is positively influenced by the reduced density, yielding 1.22, 0.62 and 0.49 W/(m·K) at the density of 1200, 900 and 600 kg/m^3, respectively. Vaou et al. [28] designed a thermal insulating geopolymer using NaOH as activator, perlite as raw material and hydrogen peroxide as a foaming agent. The thermal conductivity reduces from 0.05 to 0.03 W/(m·K) when the hydrogen peroxide dosage increases from 0.6% to 2.0% by mass of the paste, and remains constant at a further increase of hydrogen peroxide until 2.9%, while in the meantime, the compressive strength increases from 0.3 to 0.8 MPa and then remains stable. Sanjayan et al. [29] reported a lightweight aerated geopolymer concrete with a compressive strength of 0.9 to 4.35 MPa at the density range of 403–1309 kg/m^3. They found that aerating of geopolymers causes damages in stoichiometry of alkali activator and incomplete geopolymerization of fly ash particles. Zuda et al. [30] developed alkali activated lightweight composite using water glass as activator, slag as raw material and vermiculite as lightweight aggregates. The composites, with a compressive strength of 22.6 MPa at a dry density of 1918 kg/m^3, enable a fast removal of water vapour and other gaseous compounds leading to improved fire resistance properties because of their relatively high water diffusion coefficient. Kupaei et al. [31] observed an increased fly ash content causes the strength reduction due to its higher water demand and a pre-soaking of the used lightweight aggregates is observed to slightly contribute to the strength development in geopolymer lightweight concrete using oil palm shell as lightweight aggregates, fly ash as the raw material and NaOH modified Na_2SiO_3 as activator.

This thorough review indicates the potential of designing lightweight geopolymer concrete. Nevertheless, it can be seen that many available research focused on using foaming agent for the lightweight purpose, which has certain drawbacks. In overall the resulted concrete is very weak in terms of strength, for instance at a density of 900 kg/m^3, most of the concretes show a 28-day compressive strength of lower than 5 MPa, and the pore sizes of these concretes are very big, indicating a very high permeability. In addition, the very fast setting of the aerated concrete results in an un-efficient use of the raw materials. In our previous research, an ultra-lightweight PC concrete (ULWC) applying lightweight expanded glass aggregates (LWA) was developed [23]. The ULWC shows an average fresh material density of about 800 kg/m^3, 28-day compressive strengths of about 10 MPa and a thermal conductivity of about 0.12 W/(m·K) [23]. This is, to the authors' knowledge, the concrete with the lowest TC at this compressive strength and density class [32–34]. This type of concrete can be used to design more energy and ecology efficient buildings. For example, the traditional building concepts e.g., multilayer wall approach for utility construction (composed of an outside-, insulation- and indoor layer) could be replaced by a monolithic ULWC, resulting in more design flexibility for architects and reduced construction time. Therefore, motivated by this concept and starting from the reported literature, this study aims to develop an ultra-lightweight geopolymer concrete (ULWG) that would bring further the sustainable and innovative development.

In this study, an ultra-lightweight geopolymer concrete is designed and analysed, implementing alkali activated binders. A predesigned alkali activated precursor consisting of the mixture of low calcium fly ash and ground granulated blast furnace slag together with sodium hydroxide solution in combination with a superplasticizer additive is applied as the binder. A waste glass produced expanded lightweight material is applied here as lightweight aggregate. The mixture designs are modified on the basis of multiple steering parameters, whereas the previous research is taken a reference [23]. The following topics are addressed:

- The mix design approach in terms of fresh concrete stability, workability, compressive strength and TC of hardened concrete.
- Presoaking of LWA, addressing the potential particle segregation in fresh concrete;
- Fresh ULWG concrete workability and strength development characteristics, in relation to liquid binder content (L/B);
- Analyses of optimal binder contents, for optimization of compressive strength and related TC;
- Influence of LWA particle size fractions on ULWG compressive strength and TC;
- The optimal alkali dosage in the activator in terms of compressive strength;
- The effect of air entraining agent, for further reducing density and related TC;
- Element release (Si, Al, Ca, K) from raw materials (minerals) in an alkaline environment, which function as the main geopolymer network building blocks.

2. Experimental work

The ULWG was designed using a predefined geopolymer (GP) binder, supplied by Van Gansewinkel Minerals (VGM) and Cementbouw Mineralen (CB), both located in The Netherlands. This binder system (mineral precursor, alkali concentration and additive) possesses comparable mechanical properties with cement (e.g. CEM III/B 42.5 N) according to the preliminary research. The ULWG mixture design and composition calculations were performed with a specially developed model, partly based on the modified Andreasen and Andersen model which is applied in the previous research [23,35–38].

2.1. Materials

The precursor used in this study was pre-mixed by mass, 70% powder coal fly ash (PCFA) class F and 30% ground granulated blast furnace slag (GGBS). This mineral has a specific density of 2498 kg/m^3, a median particle size (d_{50}) of 15 μm (comparable to for instance CEM I) and a chemical composition as listed in Table 1(measured by XRF).

The alkali activating solution was formulated by blending water with sodium hydroxide (NaOH) which has a density of 1.36 kg/l and a molarity (M) of 11.2 to a desired molarity (2 or 3 M). Furthermore, a specially developed geopolymer additive (hereafter identified as Ad.1) was used with a fixed dosage of 3 l/m^3, supplier SQAPE Geopolymer Technology. This new type polyhydroxy carboxylate complex (PCC) improves the fresh concrete workability and related hardened material

Table 1
Chemical composition of the binder.

Oxides	SiO_2	Al_2O_3	CaO	Fe_2O_3	MgO	K_2O	SO_3	Na_2O	TiO_2	MnO	BaO	LOI
Con. (%)	52.75	18.05	12.92	5.92	3.86	2.09	1.76	1.11	1.01	0.14	0.11	1.60

Table 2
General material properties of LWA.

LWA fraction (mm)	Particle density[a] (kg/m^3)	Crushing resistance[a] (MPa)	Thermal conductivity[a] (W/(m·K))	Water absorption after 30 min (wt.%)	Water absorption after 24 h (wt.%)
0.25–0.5	540	2.9	n.a.	n.a.	n.a.
0.5–1.0	450	2.6	n.a.	5	15.1
1.0–2.0	350	2.4	n.a.	5.7	15.1
2.0–4.0	310	2.2	0.07	4.7	14.4
4.0–8.0	300	1.9	0.07	n.a.	n.a.

n.a.: not available.
[a] Provided by the supplier.

properties. Furthermore a commercial air entraining agent (AEA) (supplied by SQAPE Geopolymer Technology) was applied (3 l/m^3), which was selected from the preliminary research. The AEA introduces more micro air bubbles into the concrete which could result in a reduced density and a lower TC. In overall, water, NaOH and Ad.1 were addressed as the total liquid in this study (although it contains solids). Additionally, the concrete further consists of various fractions of highly porous expanded glass lightweight aggregates (LWA), supplied by Liaver. These are produced out of recycled glass (material properties given by producer, listed in Table 2) and possess ultra-low densities (between 300 and 540 kg/m^3) compared to normal density sand and gravel aggregates (2600 kg/m^3). These LWA contain a number of air pores (cellular structure) encapsulated in rather closed and impermeable outer shells. In addition, the differences between the size fractions resulted in different pore sizes i.e. bigger particles contain bigger pores compared to smaller particles.

Furthermore, the pores of the LWA are mostly closed (not interlinked) and the LWA possess a rather closed outer surface and cellular internal structure, as illustrated in Fig. 1.

The particles contribute to concrete with a low density, low TC and relatively high compressive strength, as demonstrated in the research on ULWC [23]. Furthermore, their glassy structure and origin, which could be amorphous, might contribute to the geopolymeric network condensation process by introducing a pozzolanic reaction [39–40].

2.2. Design methodology

As discussed in Section 1, the influence of multiple variables on the properties of the ULWG is studied. The purpose is to design a concrete with a workable fresh behaviour and a 28-day compressive strength ≥10 MPa and low TC ≤0.12 W/(m·K) in hardened state.

In addition, two different mixture design approaches were analysed, namely in the first one the solids were designed based on un-optimized packing principle and the second one on optimized packing following the modified Andreasen and Andersen model [23]. An optimized packing of the involved solids results in the lowest porosity, which contributes to an enhanced mechanical property [41]. By simply adjusting the proportion between smaller and bigger particles, in favour of the bigger particles, a mixture with an un-optimized packing is resulted which leads to a higher porosity and related lower TC, but also possibly lower mechanical property. Both approaches were applied to gain more insight of the fresh and hardened materials properties, and various mixtures were designed and tested, as listed in Table 3. In mixes GP-A1–5 90–95% 2–4 mm LWA and 5–10% 0.25–0.5 and/or 0.5–1.0 mm LWA are applied (as denoted with * in Table 3). GP-C1 is equal to GP-B2 and GP-D1 is equal to GP-C2, being listed twice only for clarification purposes.

Below a brief explanation of the various mixtures is given:

- Mixtures GP-A1–6: the properties of designs following un-optimized packing principle are evaluated.
- Mixture GP-A7: the effect of pre-soaking the LWA is analysed.
- Mixture GP-B–E: the properties of mixtures designed applying the optimized-packing approach are determined.
- Mixtures GP-B1–B4: the effect of liquid–binder (L/B) ratios is studied.
- Mixtures GP-C1–C4: the effects of binder–aggregate (B/A) ratios and LWA sizes are studied.
- Mixtures GP-D1–2: the effect of an AEA in concrete is studied.
- Mixtures GP-E1–2: the effect of NaOH molarity (M) on strength development is studied.
- Additionally, pozzolanic potential and element release of PCFA, GGBS, LWA, glass and sand within alkaline conditions are studied.

The geopolymer (GP) mixtures were prepared as follows: the water, NaOH, AEA (if any) and Ad.1 were blended. When pre-soaked LWA were applied, particles were placed in water for one hour. Afterwards, the particles were separated from the remaining water by a sieve and surface-dried with paper towels. The mineral precursor and LWA were mixed in a concrete mixer, and the liquid was added. The total mixing time was about 7 min. The rotation speed of the mixer was

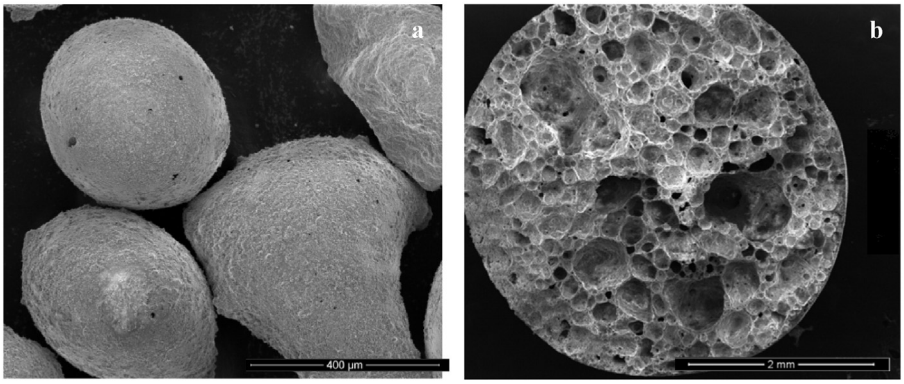

Fig. 1. SEM picture of LWA: (a) outer surface and (b) cellular pore structure [23].

Table 3
ULWG mixture designs (per m^3).

Mix	Binder (kg)	Liquid (l)	NaOH (M)	Ad. 1 (l)	AEA (l)	LWA (size in mm; amount in kg)				
						4–8	2–4	1–2	0.5–1	0.25–0.5
GP-A1–5*	450	160	3	3	–	–	*	–	*	*
GP-A6	450	180	3	3	–	–	188	–	–	17
GP-A7 (pre-soaked LWA)	450	160	3	3	–	-	184	–	30	–
GP-B1	492	140	3	3	–	70	57	33	43	29
GP-B2	492	150	3	3	–	69	57	33	43	29
GP-B3	492	160	3	3	–	68	56	32	42	28
GP-B4	492	175	3	3	–	66	54	32	41	27
GP-C1	492	150	3	3	–	69	57	33	43	29
GP-C2	492	150	3	3	–	–	127	33	43	29
GP-C3	400	140	3	3	–	74	61	36	46	31
GP-C4	400	140	3	3	–	–	136	36	46	31
GP-D1	492	150	3	3	–	–	127	33	43	29
GP-D2	457	139	3	3	3	–	118	31	40	27
GP-E1	388	174	3	3	–	–	54	68	34	122
GP-E2	388	174	2	3	–	–	54	68	34	122

lowered to a comparable speed of truck mixers when the workability was measured over time. After mixing, the fresh concrete was poured into 100 × 100 × 100 mm^3 and 150 × 150 × 150 mm^3 steel moulds and compacted for about 20 s using a vibration table. The samples were covered with plastic foil and stored at an ambient temperature of ±20 °C for 24 h, before demoulded. Subsequently, samples were covered in foil and further cured at ambient temperature of ±20 °C.

2.3. Experiments

The workability of the fresh concrete was measured in terms of flow (F) and slump (S), in accordance with EN 12350-8:2009 [42] and EN 12350-2: 2009 [43]. The initial flow without jolting was also measured, in response to the interest of practice even though this measurement is primarily intended for self-compacting concrete. The fresh concrete density was measured with a 3 litter steel cylindrical mould. The compressive strength tests were conducted to three 100 × 100 × 100 mm^3 samples of each recipe at 7 and 28 days with a controlled testing machine (Ratio-Tec). The oven-dry density was determined by drying the samples (100 × 100 × 100 mm^3) at 105 ± 5 °C, following EN 12390-7:2009 [44]. The thermal conductivity (TC) was determined twice on two oven-dried samples (100 × 100 × 100 mm^3) by a heat transfer analyser (ISOMET model 2104). The analyser applies a dynamic measurement method to determine simultaneously the volumetric heat capacity (J/(m^3·K)) and the thermal conductivity (W/(m·K)) of materials with a measurement time of about 8–16 min. 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.

The water penetration under pressure was measured on three samples of 150 × 150 × 150 mm^3 after 28 days of curing. The samples were exposed to 5 bar water pressure for 72 h and the samples were split afterwards, in accordance with EN 12390-8:2009 [45]. The total element leaching experiment was carried out on dry materials, two powders; PCFA and GGBS and three aggregates; natural sand 0-4 mm, recycled glass fraction 0–4 mm and crushed LWA 0–4 mm. All materials were mixed in a 3 M NaOH solution (mixture of demineralized (demi) water and concentrated NaOH) in total volume of 200 ml with an L/S of 5. The samples were shaken for 24 h and the eluate was filtered on 0.45 μm paper. In total 40 ml eluate was acidified with 9 ml nitric acid (70%), to a pH below 2. The method was mainly based on NEN-EN 12457-3 [46] (compliance test for granular samples). Inductively coupled plasma atomic emission spectroscopy (ICP-AES) was used for the total element analysis of the acidified eluate samples.

3. Results analysis and discussion

The influences of individual parameters on the performance of ULWG are addressed in this section. From the tested two design approaches (one based on optimized particle packing and another one not), the overall performance of the un-optimized packing approach is not convincing and mixtures were found instable. The results are described in Section 3.1. The results of the mixtures designed based on the optimized packing principle possess stable and predictable performances. This mixture design approach is further applied to analyse the effects of the various parameters (L/B ratio and binder/LWA ratio influences, LWA fraction size performance, AEA use, activator alkalinity) on the performance of ULWG.

3.1. Mixtures based on an un-optimized particle packing approach

In preliminary tests the maximum dosage of large LWA particles (fraction 2–4 mm) was determined. Based on this measurement, mixture design is further constructed by filling the remaining voids with fractions of LWA 0-1 mm under a fixed binder dosage. To assess the effect of such an un-optimized packing approach, the aggregates consist of 90–95% large (i.e. 2–4 mm) and 5–10% small (i.e. 0–1 mm) size aggregate fractions. When the concrete was produced (mixture GP-A1–A5, Table 3), the overall workability was poor and almost zero slump was encountered. Furthermore, the system tends to be very sensitive to particle segregation when increasing the liquid from 160 to 180 l/m^3. The additional fresh concrete compaction was insufficient due to firstly, at a low liquid content (e.g. 160 l/m^3) the concrete was too stiff to generate an optimal compaction level and secondly, with a higher content (180 l/m^3) it shows particle segregation making the mixture unstable. The final concrete contains many large air bubbles, both in the concrete matrix and on the surface. A picture of the cross section of an ULWG by an un-optimized packing approach is shown in Fig. 2a.

The samples have the following performances: fresh density of around 830 kg/m^3, oven-dry density of around 710 kg/m^3, 28-day compressive strength of 8–10 MPa, thermal conductivity of 0.153–0.232 W/(m·K) and water penetration of 16–26 mm. The thermal conductivity is quite low at this strength and density class compared with ordinary building materials due to the cellular internal structure of the LWA (see the SEM pictures in Fig. 1). However, in overall, the performance of the mixtures designed based on an un-optimize packing applying dry LWA are found to be insufficient (i.e. poor compaction and a higher thermal conductivity compared to the ULWC based on cement) and therefore this design approach is not further tested.

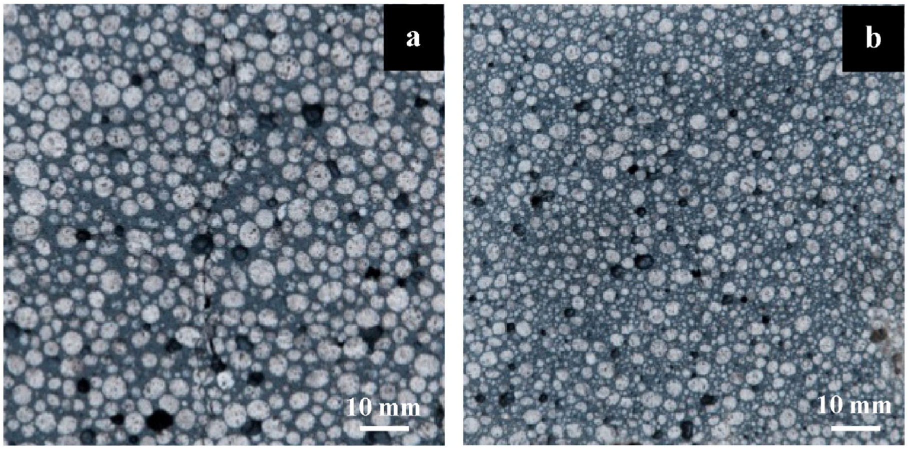

Fig. 2. Cross section of ULWG: design using: un-optimized (a) and optimized (b) particle packing approach.

3.1.1. *Effect of pre-soaking LWA*

The water absorption experiments show that the aggregate absorbs more liquid over time compared to natural sand of gravel (Table 2), even though the SEM pictures in Fig. 1 show a rather closed outer surface.

As stated, when dry LWA are applied in un-optimized packing mixtures, large air bubbles occur or segregation takes place. Applying water pre-soaked LWA may avoid segregation and may contribute to more stable mixture and better compactible concrete. By pre-soaking, less paste will be absorbed from the fresh concrete which strongly affects the workability in time as LWA are already partly saturated. As a consequence, the workability may improve since more liquid is then available to provide workability. However, it has to be taken into account that an effective pre-soaking of LWA is difficult to achieve due to the very small particle sizes and low density (±300–500 kg/m^3). Additionally, from trials it is observed that the dry LWA may absorb a large amount of liquid. As a rule of thumb it can be assessed, that increasing the LWA particle size means a higher absorption. In mixture GP-A7 (Table 3) it is observed that the pre-soaked LWA in the mixture generated more reliable characteristics compared to the non-presoaking mixtures in terms of stability, compaction, air content, porosity and particle distribution. Still its overall fresh and hardened state performances are poorer than expected. The concrete has the properties: fresh density of 850 kg/m^3, oven-dry density of 687 kg/m^3, 28-day compressive strength of 9.1 MPa and TC of 0.133 W/(m·K).

3.2. *Mixtures based on an optimized particle packing approach*

The mixture design methodology based on an optimized particle packing approach has been addressed in detail in the previous research [23,35–38]. The mixture designed based on the optimized packing approach (GP-B–E, Table 3) show overall better and stable fresh and hardened concrete properties in terms of stability, compaction and porosity. A picture of the cross section of a hardened mixture is illustrated in Fig. 2b. It is shown that less air bubbles are present in the mixture, the particles are distributed more homogeneously in the matrix and the small LWA fill up the area between the bigger particles which results in a more stable specimen.

3.2.1. *Effect of liquid/binder ratio*

Within common concrete practice, the modification of fresh concrete workability is mainly influenced and controlled by three parameters, including total solid particle packing, liquid content and admixtures. In this study the influence of liquid content on the fresh ULWG workability is mainly addressed by varying the L/B ratio from 0.28 to 0.36 (recipes GPB1–4, Table 3). The workability is analysed by measuring slump, initial flow without jolting and flow with jolting, see Fig. 3 (Deviations are within 5%).

Increasing the L/B ratio and the related mixture liquid content from 140 to 175 l/m^3 (based on own preliminary research) strongly affected the fresh concrete performances, with the fresh mortars changing from very stiff to mixtures with self-levelling properties. At the liquid dosage of 140 l/m^3 (L/B 0.28), the mortar was very stiff (zero slump), while at the 150 to 160 l/m^3 (L/B 0.3-0.33) the workability was relatively high, with a slump ≥25 cm and flow ≥55 cm. This is in line with common practice and previous research, showing a workability increase when L/B ratio rises. The overall result indicates that a significant range between zero and high workability is caused by a small extra amount of liquid. This makes the ULWG concrete potentially sensitive to liquid content. Furthermore, the workability in time up to 60 min strongly

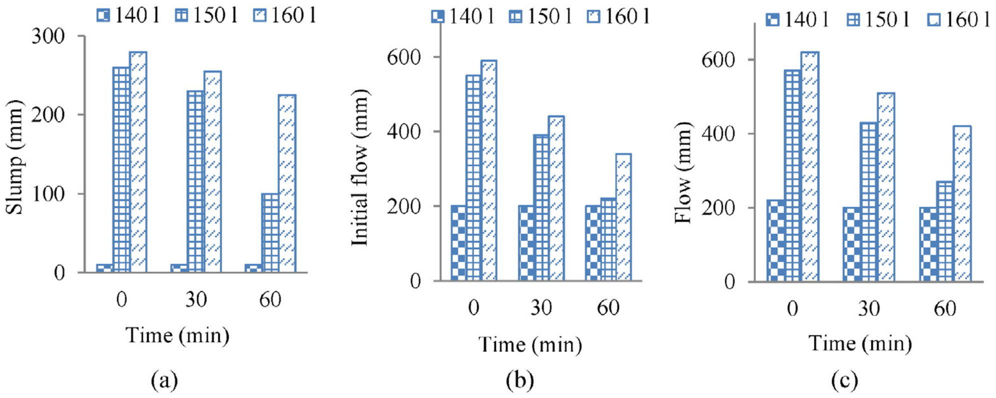

Fig. 3. Workability of fresh ULWG: (a) slump, (b) flow without jolt, (c) flow after 15 times jolts.

Fig. 4. Workability of mixture GP-B2: (a) right after mixing, (b) one hour after mixing.

declines due to the high LWA liquid absorption (described in Section 3.1.1). Fig. 4 shows the workability of recipe GPB-2 with 150 l/m^3 of liquid direct after mixing (a) and at one hour (b). It is shown that the slump decreases significantly in one hour. Furthermore a minor detail to mention, within the ULWG experiments fresh concrete temperatures varied between 20 and 24 °C and remained constant up to 60 min. The preliminary research also showed that the used GP mineral binder produces a temperature development of up to around 40 °C. This is fairly low compared to that of normal PC based mixtures and their relatively high temperature development of ≥60 °C.

The determined calculated, fresh and oven-dry mixture densities are illustrated in Fig. 5a. Deviations are within 5%. In overall, the calculated and measured fresh concrete densities are relatively similar. The increase of fresh density in time is related to the liquid absorption of the LWA in time, as indicated by Table 2. This increases the LWA in mass, without changing its volume. However, the related oven-dry densities overall strongly decrease up to 15% when more liquid is used in the mixture, caused by evaporation.

Increasing the total liquid content (or higher L/B ratio) of the mixture results in a higher water permeable porosity which negatively affects the compressive strength. As a consequence, in the experiments the strength is reduced, up to 17%, when the liquid content increases from 140 to 150 l/m^3 to 175 l/m^3 (see Fig. 5b). This is in line with the previous research which shows that when the L/B ratio is too high, an optimal strength development is not generated [47–50]. Besides the strength, the thermal conductivity was also measured, and the results are presented in Table 4.

In overall it is illustrated that the TC decreases up to 17% (as compressive strength) when the L/B ratio is increased. As explained, at a higher L/B, more liquid evaporates in oven-dry state which increases the voids in the concrete which favours the TC. This is also in good agreement with the previous research, stating that TC decreases at a lower density [34,51]. The overall relations between the compressive strength, thermal conductivity and L/B ratio (with 140, 150, 160 and 175 l liquid per m^3 mixture, respectively) are summarized and plotted in Fig. 6.

Finally, the water penetration under pressure of the hardened ULWG is tested and the results are listed in Table 4. In overall, the water penetration is between 12 and 17 mm, showing that the concrete is relatively low-permeable in structure even when it contains extremely high amount of porous LWA.

3.2.2. Effect of binder/aggregate ratio and LWA size

To further optimize the ULWG concrete, two additional design parameters were investigated. Firstly, the binder/aggregate (B/A) ratio was studied. Decreasing the absolute binder content could reduce the TC as a consequence of the reduced concrete density. As stated, the particle density of the binder (2500 kg/m^3) is about 6 times of the LWA (average 400 kg/m^3) and this is therefore the most dominating parameter affecting the concrete porosity. Secondly, the effect of the LWA particle size was tested. Replacing the 4–8 mm LWA (crushing resistance of 1.9 MPa) by 2–4 mm LWA (crushing resistance of 2.2 MPa) may possibly further increase the ULWG compressive strength.

3.2.2.1. Effect of binder/aggregate ratio. In mixtures GP-C1–2, binders of 492 kg/m^3 and in mixtures GP-C3–4, 400 kg/m^3 are used, as listed in Table 3. The experiments show that when applying a lower B/A ratio, the fresh concrete workability (slump and flow) significantly degrades. Applying less binder and more LWA leads to an increased amount of the liquid absorbed by the LWA.

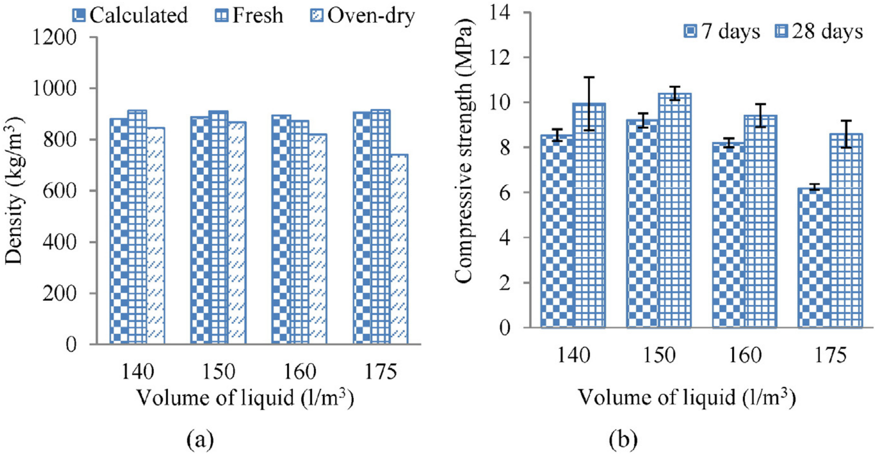

Fig. 5. (a) Density, and (b) compressive strength of ULWG versus liquid/binder ratios.

Table 4
ULWG properties with various liquid dosages.

	GP-B1	GP-B2	GP-B3	GP-B4
Thermal conductivity (W/(m·K))	0.172	0.174	0.159	0.144
Water penetration (mm)	12.3	17.0	16.0	n.m.

n.m.: not measured.

From the mixtures it is observed that with a binder reduction of about 25%, the fresh density declines from 949 to 808 kg/m^3 (17%). Consequently, the 28-day compressive strength declined up to 25% (see Fig. 7).

A lower density causes a lower compressive strength [52–53]. Nevertheless, the lower B/A ratio significantly contributes to a decrease of the TC of the mixtures with 4.0–8.0 LWA (0.174 to 0.074 W/(m·K)) and without 4.0–8.0 LWA (0.127 to 0.070 W/(m·K)), as shown in Table 5).

This is in line with the previous research, which stated that the thermal conductivity decreases from about 1.75 to 0.16 W/(m·K) when lowering the density from 2300 to 400 kg/m^3 [51–53]. Compared to the most optimal TC of ULWC (PC based) which was 0.12 W/(m·K) [23], the ULWG illustrates a potential to generate an extremely low TC (0.07 W/(m·K)) at a moderate compressive strength of 8 MPa. Furthermore, it is observed that the water penetration under pressure is higher at a lower B/A ratio, illustrated in Table 5. In this case, lowering the binder content leads to a decreased material density and higher porosity, which result in a higher water permeability [54]. Nevertheless, the results are still within the acceptable range.

3.2.2.2. Effect of LWA size. In mixtures GP-C1 and GP-C3, LWA of 4.0–8.0 mm are applied and in mixtures GP-C2 and GP-C4, the 4.0–8.0 mm LWA are replaced by 2.0–4.0 mm LWA, as listed in Table 3. It is indicated that the workability significantly decreases when the 4–8 mm LWA are replaced by 2–4 mm LWA. This is attributed to the fact that the water demand of smaller LWA particles is higher than bigger particles, due to the increase of the specific surface area [55].

The replacement of larger particles by smaller particles has no significant effect on the calculated, fresh and oven-dry densities. Nonetheless, it positively influences the compressive strength and TC. The strength increases up to 11% when the volume of 4–8 mm LWA is replaced by 2–4 mm LWA, see Fig. 7. As stated, this can probably be attributed to the higher crushing resistance of the smaller particles (given in Table 2). The TC declines with 27% from 0.174 to 0.127 W/(m·K) when 492 kg/m^3 binder is applied (Table 5). This is in line with the previous research, i.e. the thermal conductivity reduces when smaller particles are applied as a consequence of the air pores (in the LWA particles) which can be distributed more homogeneously in the mixture [22]. When 400 kg/m^3 binder is applied, the effect of LWA size is less prominent, with only a very slight decrease from 0.074 to 0.07 W/(m·K), Table 5. The TC of the mixture with 4–8 mm LWA is already almost equal to the TC of 2–4 mm LWA (0.07 W/(m·K), Table 2). Therefore, it is most likely that the replacement of the LWA will positively affect the strength, while the TC remains similar.

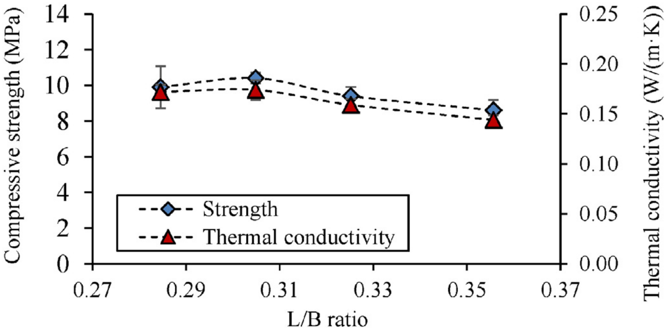

Fig. 6. 28 day compressive strength and thermal conductivity versus L/B ratio.

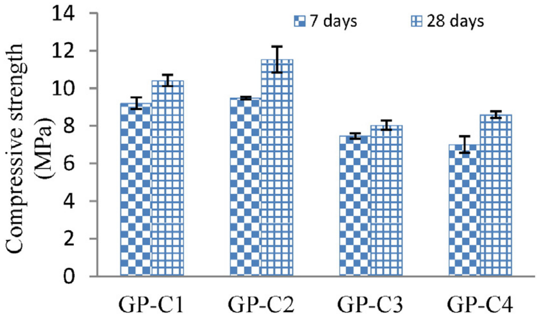

Fig. 7. Compressive strength of ULWG with different binder contents or LWA fractions. Binder and LWA fraction size respectively: GP-C1: 492 kg; 0.25–8 mm, GP-C2: 492 kg; 0.25–4 mm, GP-C3: 400 kg, 0.25–8 mm, GP-C4: 400 kg, 0.25–4 mm.

Nevertheless, it is noteworthy that here it is also observed that the TC increases (0.07 to 0.14 W/(m·K)) when an excessive amount of small particles is applied (44 instead of 8 wt.% 0.25–0.5 mm LWA and 20 instead of 63 wt.% 2–4 mm LWA, Mixes GP-E and GP-C4, see Table 3). This indicated that a certain balance between small and big particles to achieve the lowest TC is desired.

The water penetration under pressure decreases when the smaller particles are applied, see Table 5 (GP-C2 compared to GP-C1 and GP-C4 compared to GP-C3). This is in line with the previous research which reported that the water permeability increases when the interconnectivity of the pores increases, which is the case when larger particles are applied [56].

3.2.3. Effect of air entraining agent

Air pockets (macro to micro air bubbles) within concrete are known as a crucial parameter to influence the density, strength and TC, i.e. the insulation potential. The influence and performance of a proper working air entraining agent (AEA) is analysed whereby mixture GP-D1 (promising mixture) is used as a reference. Within this recipe, an AEA with a dosage of 3 l/m^3 is added (mixture GP-D2). The density of the mixture declined with 7.7% from 949 (GP-D1) to 881 (GP-D2) kg/m^3 by entrapped air and increased porosity (Fig. 8, deviations within 5%), which is in line with the previous research [34,51,57].

In other words, about 8% of air is formed in the fresh concrete. As a result, the TC decreased with 15% compared to the reference, from 0.127 to 0.108 W/(m·K) (see Table 6), which is in agreement with previous research [34,51–53].

Despite the positive influence of the air entraining agent on the density and TC, 16% loss of compressive strength is observed (11.5 to 9.7 MPa; Fig. 9), which is also in line with the previous research [52–53]. The water penetration under pressure rises from 8.3 to 22.7 mm for the mixture without and with AEA (Table 6). In the previous research it is observed that with a lower density (which is the case in the mixture with AEA) an increase of water permeability is observed, possibly due to the capillary effect and the interconnection between air pores [54].

Table 5
ULWG properties with various binder/aggregate ratios and different LWA particle sizes fraction.

	GP-C1	GP-C2	GP-C3	GP-C4
Thermal conductivity (W/(m·K))	0.174	0.127	0.074	0.070
Water penetration (mm)	17	8.3	17.3	12.3

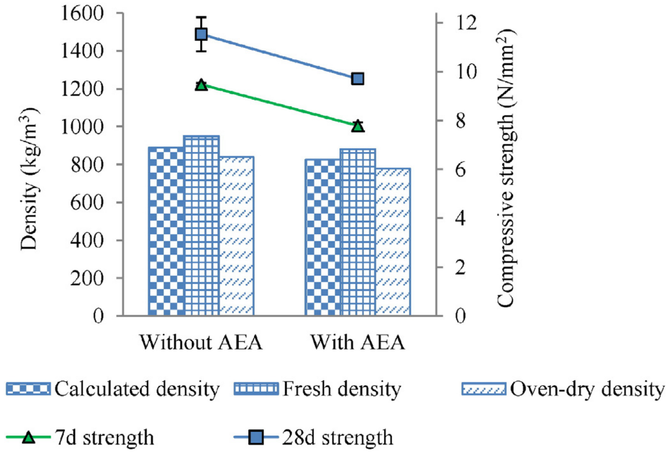

Fig. 8. Densities and compressive strengths of ULWG with and without AEA.

3.2.4. Effect of NaOH concentrations

The concentration of NaOH is of great importance affecting the AAM properties as for example compressive strength development [58–59]. Compared to the literature, in this study already a relatively low concentration of activator of 3 molar (M) is used. To analyse its influence on the performance of ULWG and in addition to optimize the utilization, further lowering of the alkalinity to 2 M (GP-E2) is tested as listed in Table 3.

Both fresh concrete mixtures did not show clear differences, since both had a very poor workability (no slump). Additionally, the density, TC and water penetration under pressure were not significantly deviated. Nevertheless, the 28-day compressive strength at 2 M compared to 3 M significantly increased with 36% from 6.6 to 9 MPa (Fig. 9).

This is in contrast with literature, stating that the compressive strength increases towards an optimum when the NaOH molarity increases from 2–3 to 4–6 M (from 18 MPa at 3 M NaOH to 25 MPa) [59]. The strength development and polycondensation of the network is mainly affected by a number of factors, including the total element composition silicon (Si), aluminium (Al), calcium (Ca) and sodium (Na), their amorphous or mineral state and the ratio of the binder and aggregates [60–61]. As the predefined GP binder concept used within this study has shown the most effective in strength development at 3–5 M NaOH (tested in normal density concrete mixtures based on 400 kg binder in combination with sand and gravel). The strength increase of the ULWG at 2 M may be explained by the amorphous glass structure of the LWA, which is mainly composed of the elements Si and Na. These amorphous LWA will be more reactive in comparison to crystalline structured sand and gravel. If so, the extra dissolution of these elements from the LWA enhances more network growth and reaches its optimum Si:Al, SiO_2:NaOH and SiO_2:Na_2O ratio and related higher compressive strength at a lower molarity [62–63]. It should be noted that in the preliminary research it was found that when the molarity NaOH decreases from 2 M to 1 M, the 28-day compressive strength was strongly reduced. This is most likely caused by the fact that a certain NaOH threshold concentration is needed to facilitate and activate the system, in order to develop a proper mechanical property.

Table 6
ULWG properties with and without AEA.

	GP-D1	GP-D2
Thermal conductivity (W/(m·K))	0.127	0.108
Water penetration (mm)	8.3	22.7

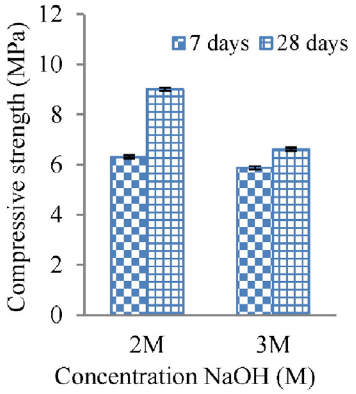

Fig. 9. Compressive strength of ULWG with different NaOH concentrations.

3.3. Element release of raw materials

Alkali activated or geopolymer network structures are mainly built out of the elements Si, Al, Ca, Na, K and Mg [3,52,64]. Their abundance and release in time results in network condensation and final structures with related material properties. The potential release of some of these elements from the used aggregates could affect the GP network formation. The LWA chemical composition is 71% Si, 13% Na, 8% Ca and 2% Al, in combination with its amorphous structure (instead of a crystalline structured sand or gravel), could act as a mineral precursor and/or alkaline activator. With a cascade leaching test within a high alkaline environment (3 M NaOH solution) the element release of LWA is measured. The results are compared to other raw concrete materials; powder coal fly ash (PCFA), ground granulated blast furnace slag (GGBS), glass and sand, as illustrated in Fig. 10.

Na could not be accurately measured due to the high content, related to the NaOH. Mainly a high release of Si is shown by the LWA and PCFA, which is much higher compared to that of glass and sand. This is most likely because both materials are mainly composed of reactive glassy silicon structures. Furthermore, LWA has a moderate release of element Al and K and almost no Ca. Although, the LWA only consist of 2 wt.% Al (23% in PCFA), its relatively high abundance within the eluent indicates that its initial structure is relatively soluble. In overall, the results show that the alkaline environment stimulates the LWA and PCFA particle solubility and element release whereas the glass and natural sand show much lower releases. Since the mixtures GP-E1–2 were composed of 388 kg/m^3 GP binder (i.e. 116.4 kg/m^3 PCFA) and 278 kg/m^3 LWA, the high amount of LWA may provide the GP system with significant amounts Si, Al and maybe Na building block elements. The more optimal Si:Al, SiO_2:NaOH and SiO_2:Na_2O ratios are therefore possibly achieved at a lower NaOH molarity compared to ordinary GP concrete (based on sand and gravel aggregates), as suggested in Section 3.2.4.

Finally, it can be proposed that the matrix between solid LWA and the GP gel network is effectively interconnected, resulting in a higher strength and an improved performance. This is supported by the idea

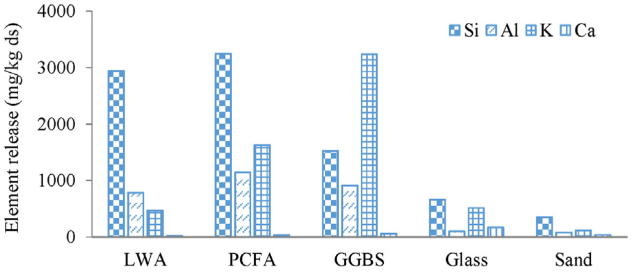

Fig. 10. Element release of raw materials in alkaline environment.

that due to element release of the LWA or in other words dissolution of its outer surface, network formation also starts at the outer surface generating a more effective interconnection between solid particles and formed gel structures. It is worthy to mention that no negative effect on the compressive strength of the developed concrete is observed.

3.4. Overall analysis and discussion

An overall analysis of the obtained results from this study is performed by addressing the relationships between various parameters, namely density, compressive strength and TC. They are plotted and compared with available data of ultra- and lightweight PC based concretes, since no research has been reported on ultra-lightweight GP concretes to the authors' knowledge [23,65–69].

3.4.1. Compressive strength versus density

The 28-day compressive strength versus the apparent density is plotted in Fig. 11a.

A trend is shown between both parameters, which can be described as the quantitative amount LWA in a mixture mainly influences the mixture porosity and density, affecting the related strength. When less LWA is added and replaced by the binder in the mixture (maintaining an equal volume) porosity decreases and density and related strength increases. Deviations from the trend line of Fig. 11a are caused by the variations in composition of the mixtures like LWA fractions and L/B ratio [47–50]. In addition, it is found that the results obtained in this study are in line with the indicated trend of lightweight concrete, obtained from data points from previous research, shown in Fig. 11b.

3.4.2. Thermal conductivity versus density

There is a clear trend found between the TC and density which is controlled by the material density and related total binder to aggregate ratio, illustrated in Fig. 12.

A strong decline of the TC is noticed when ULWG density drops. This is mainly affected by decreasing the binder content will result in a lower material density which favours an increase in porosity and a reduced TC. All data points (Fig. 12) below the trend line are generated from mixtures with a low binder content (400 kg/m^3 instead of 492 kg/m^3) and/or 2-4 mm instead of 4-8 mm fraction LWA.

3.4.3. Thermal conductivity versus compressive strength

In line with the previous research, it is shown that the TC rises as a function of compressive strength, as illustrated in Fig. 13 [52,53,70]. This effect on the development of the TC is less strong compared to material density, as shown in Fig. 12. The reduction of TC is mostly affected by two parameters; primarily by a reduced binder content or so-called binder/LWA ratio (400 instead of 492 kg/m^3 binder in a mixture), and secondly to a lower extent by the use of LWA faction and replacement of 4–8 mm by 2–4 mm LWA.

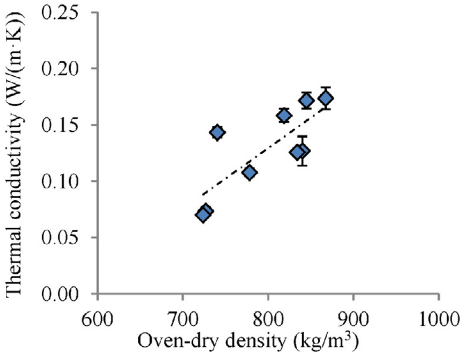

Fig. 12. Thermal conductivity versus density.

Additionally, the obtained TCs from the ULWG are significantly lower (at an equal compressive strength level) compared to the other PC based concrete from literature, as illustrated in Fig. 13b.

This can be partly explained by the used raw materials and their properties. Firstly, the LWA used in this study and in the study of ULWC is low in density and relatively high in crushing resistance. For example the aggregates applied in literature are expanded glass and foam having a lower crushing resistance [67–68] or expanded clay with a higher density [65].

Furthermore, some of the ULWG mixtures produced a significantly lower TC compared to the reference ULWC, even when a comparable binder content was used [23]. This difference can be explained by that a reduction in TC is reached when part of the PC binder was replaced by fly ash or blast furnace slag. The reduction of the binder density is the main reason for the decreased TC [71]. In addition, compared to PC these binders are significantly lower in Ca and higher in Si and due to their amorphous structure, a lower thermal conductivity is obtained [71]. Related to the GP binder within the ULWG (composed out of PCFA and GGBS), this could be a possible explanation for the lower TC of the ULWG compared to that of the ULWC [51,71].

3.4.4. Comparisons with other building materials

The designed ULWG showed very promising thermal properties compared to other traditional building materials, especially at a comparable strength class. For example, ordinary concrete with a compressive strength ≤40 MPa has a high TC of around 1.7 W/(m·K) [72]. ULWC has a TC of 0.12 W/(m·K) at a strength of 10 MPa [23]. Cement based wood wool boards have a TC of 0.08 W/(m·K), however are not designed for load bearing purposes. Stone wool and glass wool products have a TC

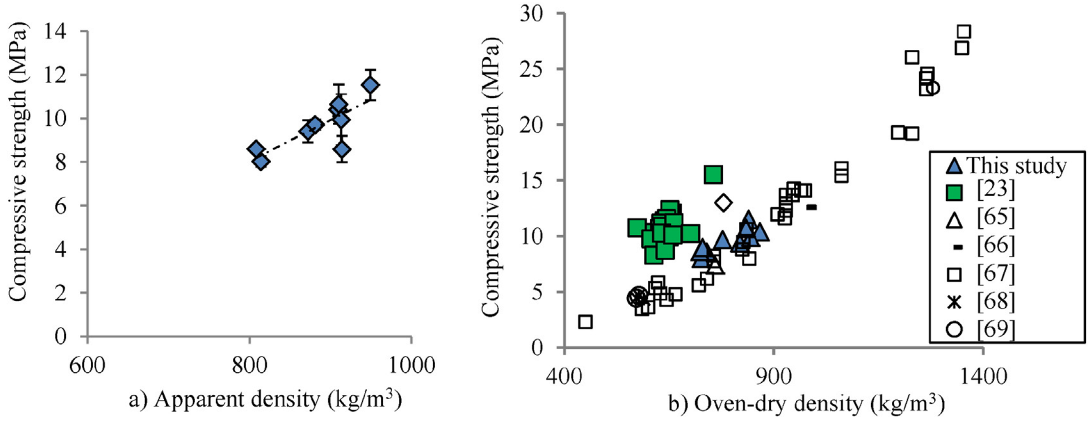

Fig. 11. Compressive strength versus (a) fresh density, (b) oven-dry density.

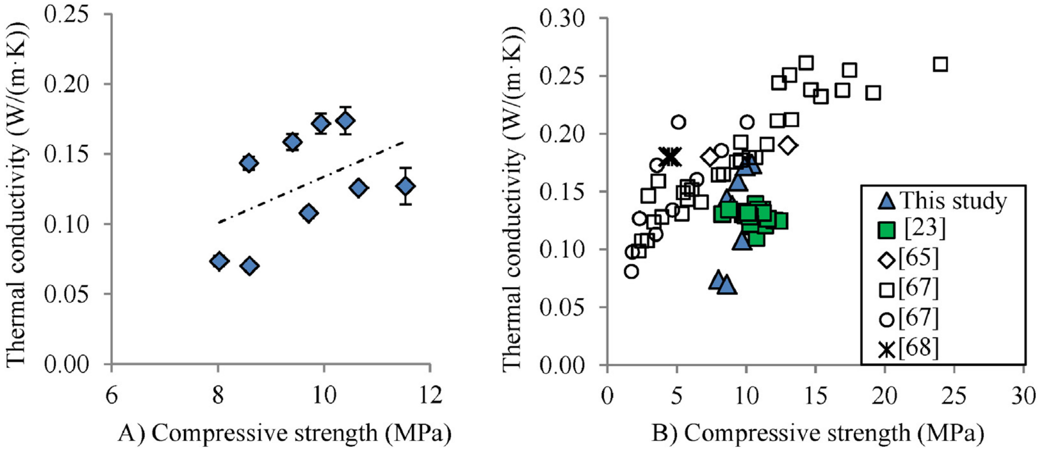

Fig. 13. Thermal conductivity versus compressive strength of (a) ULWG and (b) concrete.

of respectively 0.035 and 0.040 W/(m·K), respectively, which however are also not designed for load bearing purposes [73–74]. This highlights the excellent properties of ULWG with a TC of 0.07 W/(m·K) and a 28-day compressive strength of 8 MPa.

4. Conclusions

This study addresses the design, production and performance evaluation of ultra-lightweight geopolymer concrete (ULWG), a potential load bearing and thermal insulating building material. The applied geopolymer system has, compared to PC and other GP binders, superiorities in terms of sustainability, favoured by the use of the industrial by-products mineral as binder raw materials and relatively low NaOH activator molarity (2–3 M). With a compressive strength of 10 MPa, a very low thermal conductivity of 0.108 W/(m·K) is resulted by the developed ULWG, and the thermal conductivity even further reduces to 0.074 W/(m·K) at a compressive strength of 8 MPa. By understanding the influences of all the controlling parameters and related material properties, a fully optimized and highly effective product is resulted. Based on the performed study, the following conclusions are reached:

- *Mix design approach*: mixes designed applying an optimized packing approach gain improved mixture stability and workability with an effective porosity and lower TC.
- *Liquid/binder* (*L/B*) *ratio*: increased liquid dosage (140 to 175 l/m^3) and related higher L/B ratio affect the workability significantly (slump from 10 to 280 mm) and lower the oven-dry density, 28-day compressive strength and TC with 15–17%, due to the increased porosity.
- *Binder/aggregate* (*B/A*) *ratio*: the B/A ratio greatly influences the fresh and hardened ULWG performance. A higher ratio increases the 28-day compressive strength (up to 25%) and lowers the water penetration under pressure. Nevertheless, both fresh density and TC increase up to 17% and 58%, respectively.
- *LWA particle size*: when LWA of 4–8 mm are replaced by LWA of 2–4 mm, the ULWG performance is mainly positively affected. The 28-day compressive strength increases (up to 11%) and TC decreases (up to 27%).
- *Air entraining agent* (*AEA*): the introduction of an AEA optimizes the mixture performance by further lowering the density (7.7%) and TC (15%). This is initiated by the created micro air bubbles. Nevertheless, a 16% loss of compressive strength is observed.
- *Alkali activator concentration*: A higher compressive strength (36%) of ULWG is generated by lowering the alkali concentration from 3 to 2 M.
- *Element release of LWA*: within an alkaline environment, a certain element release from the applied LWA is shown, mainly Si and in lesser extent Al and K as network building element. Nevertheless, the effect on the compressive strength of the concrete is negligible.

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