Effects of an eco-silica source based activator on functional alkali activated lightweight composites

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Effects of an eco-silica source based activator on functional alkali activated lightweight composites

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HIGHLIGHTS

- Eco-friendly functional alkali-activated lightweight composites are developed.
- Effect of eco-silica source on alkali activation and carbon footprints is assessed.
- Effect of alkali activator on acoustical property is studied.
- Relationship between thermal conductivity and density is derived.
- Effect of alkali content on compressive strength is evaluated.

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ABSTRACT

In this paper, alkali activated slag-fly ash lightweight composites with moderate densities between around 1200 and 1500 kg/m³ are prepared and characterized. An eco-olivine nano-silica is applied to prepare sustainable silicate based activators to replace commercial sodium silicates. Na₂O contents of 2.0, 3.5 and 5.0 wt% are investigated in order to reach a suitable balance between performance, costs and application. The results show the positive effect of density and Na₂O content on strength, while strength increment between Na₂O dosage of 3.5% and 5.0% is limited. The reduction of Na₂O content shows a dramatic delay of the reaction process up to around 3 d, but shows negligible effect on the typical Si-O bonds. An increased Na₂O content benefits the formation of reaction products, including the contents of hydro-talcite and carbonates. Besides, the thermal conductivity and acoustical absorption properties of the lightweight products are characterized; the phase transition zones between lightweight aggregate and binder matrix are evaluated by SEM. The calculation on the carbon footprint shows an evident advantage of using alkali activated materials to replace Portland cement, also the utilization of olivine nano-silica further reduces the carbon emission of the activator by around 25%.

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1. Introduction

Lightweight concrete has been widely applied as both structural and non-structural components in a wide range of weights and strengths for various applications [1,2], due to its properties such as low density, good thermal insulation and fire resistance [3]. Lightweight concrete can be categorized into three grades in general, depending on the density: low density ones with densities lower than 800 kg/m³, moderate ones with densities between 800 kg/m³ and 1400 kg/m³ and structural ones with densities between 1400 kg/m³ and 2000 kg/m³ [4]. In addition, Portland cement is commonly used as binding material for lightweight con-

crete, but its production is responsible for around 7% of the global carbon emissions and high energy costs [5,6]. In order to reduce the negative environmental impacts, the development of sustainable alternatives such as alkali activated materials has been investigated because of the excellent mechanical properties, durability, thermal resistance together with low energy and carbon costs [7–9]. Some efforts have also been spent on applying alkali activated materials in producing lightweight products; including for instance the study of design methodologies of ultra-lightweight geopolymers by applying the particle packing approach, the effect of key factors such as suitable ratios between binder, activator and aggregates [10]; the development of lightweight geopolymers with foaming agent especially for thermal insulating properties [11]; investigating the relations between density, mechanical properties and thermal conductivity of geopolymers with medium to low

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densities [12–14]; the utilization of different alkali binders and unconventional aggregates [15–17].

In terms of the alkali activated binders, they are usually produced by mixing alkaline activator solutions with solid precursors, and two typical binding systems can be classified: one is the Si and Ca enriched system, having a chain structured C-A-S-H type gel as main reaction product [18]; another is the Si + Al system, the reaction product is mainly a three-dimensional N-A-S-H network [19]. Recently, the blended alkaline system, which is a mixed binder system that combines two types of typical binders together, has received increased attention due to the modified properties such as setting times, workability, shrinkage, mechanical properties and durability [20–23]. Synthesizing factors that are related to key performances of blended system such as activator and raw materials' parameters, curing conditions on reaction kinetics, gel characters, mechanical properties and durability issues have been widely studied [24–28]. Those understandings make this type of material a brighter future to applications.

In alkali activated materials, the activator plays an important role on the activation process by providing necessary alkalinities for the decomposition of raw materials and formation of reaction products. The commonly used activators are alkali hydroxides, silicates, carbonates, sulfates, aluminates or oxides; and it is widely accepted that silicate based ones ($M_2O \cdot nSiO_2 + MOH$) usually result in ideal mechanical properties and relatively low porosity [18,29]. The silicate based activator exhibits advantages by providing the alkaline environment continuously and moderately, also the additionally provided silicates from the activator will participate into the reaction products and lead to a refined microstructure [30], and sufficient content of M_2O promotes the activation efficiency while exceeding dosage show limited further contribution. However, the primary drawback of silicate based activators is its negative environmental impact. The commercial process of sodium silicate production includes the melting of sodium carbonate and quartz sand around 1400–1500 °C with carbon release of above 400 kg/ton [31]. In order to achieve a more eco-friendly binder system, a green olivine nano-silica is applied as alternative silica source to prepare activator solutions in this study. The olivine nano-silica is produced by dissolving olivine stone in acid under temperatures lower than 95 °C, and this process shows advantages in carbon emission, energy consumption and total costs [32]. Therefore an alkali activated product with further reduced overall environmental impacts can be expected.

The objective of this study is to evaluate an eco-silicate source based alkali activator in designing lightweight composites, using blended alkali activated binder. The binder materials are chosen to be ground granulated blast furnace slag and class F fly ash due to their worldwide availability and relatively stable properties, and the lightweight aggregate is based on a natural expanded silicate. The lightweight composites are designed with the aim to have moderate densities together with excellent mechanical, thermal and acoustic properties. Special focuses are paid on the efficient usage of activators and the effect of alkaline conditions on the lightweight aggregate, because the silica based, porous structure of lightweight aggregate would show an interaction with the pastes due to either dissolution of silica or absorption of activator solution. The effect of activator dosages, addition of lightweight aggregates on early age reaction, gel structure, and aggregates-binder transition zones are investigated by using isothermal calorimeter, Thermo-gravimeter (TG), Fourier transform infrared spectroscopy (FTIR) and Scanning electron microscopy (SEM) analysis. The mechanical properties, thermal conductivity and sound absorption properties are evaluated and the effect of the alkali content on these properties is investigated.

2. Experiment

2.1. Materials

The powder raw materials applied in the present work are blast furnace slag and Class F fly ash, and their major chemical compositions are shown in Table 1. Commercial lightweight aggregates produced from natural expanded silicate with three particle sizes are applied: 0.5–1 mm, 1–2 mm and 2–4 mm, with particle densities of 600, 550 and 500 kg/m³, respectively. CEN standard sand is also used as fine aggregate. The SEM pictures of the lightweight aggregates' surface and internal structure are provided in Fig. 9(A and B). Analytical sodium hydroxide and laboratory prepared olivine nano-silica (19.04% SiO₂ and 80.96% H₂O by mass) were used to produce alkali activators. The olivine nano-silica was produced by dissolving olivine in acid at low temperatures, followed by a washing and filtering process [33,34]. Desired activator parameters were obtained by mixing calculated dosage of sodium hydroxide, olivine nano-silica and distilled water.

2.2. Sample preparation

Three levels of equivalent Na₂O content were used in this study: 2.0%, 3.5% and 5.0% (by mass of the binder). A fixed activator modulus of 1.4 and a slag/fly ash mass ratio of 8/2 were used based on the previous experiences [35,36]. Mixtures are designed to have oven dry densities between 1200 and 1500 kg/m³, i.e. within the density class of D1,4 and D1,6 according to EN 206-1 [70]. The detailed mix proportions are presented in Table 2, for instance, sample with the label of D15-5.0 means it was designed to have a density level of 1500 kg/m³ and a Na₂O content of 5.0%. Specimens were prepared and poured into molds of 40 × 40 × 160 mm³, then covered with a plastic film to prevent the moisture loss. All specimens were demolded after 24 h of curing and cured at room temperature and a relative humidity of 95% until testing age.

2.3. Testing methods

The compressive strength was determined according to EN 196-1 [37]. Samples were tested at the ages of 7 and 28 d, respectively. The early age hydration heat release was investigated by an isothermal calorimeter with TAM Air, Thermometric. Measurements were carried out for 72 h under a constant temperature of 20 °C. Fourier transform infrared spectroscopy (FTIR) study was conducted in a Varian 3100 instrument with a resolution of 1 cm⁻¹, and the applied wavenumbers were between 4000 and 600 cm⁻¹. The thermal analyses were performed in a STA 449 F1 instrument; the testing temperatures were up to 1000 °C with an increment of 5 °C/min, protected by nitrogen gas. The thermal conductivity (k) and the mass heat capacity (c) were measured by

Table 1
Major chemical composition of slag and fly ash.

Oxides (wt.%)	FA	GGBS
SiO ₂	54.62	34.44
Al ₂ O ₃	24.42	13.31
CaO	4.44	37.42
MgO	1.43	9.89
Fe ₂ O ₃	7.21	0.47
Na ₂ O	0.73	0.34
K ₂ O	1.75	0.47
SO ₃	0.46	1.23
LOI	2.80	1.65

Table 2Mix proportions of alkali-activated slag-fly ash composites (kg/m^3).

Sample	Precursors		Activator			Sand	Lightweight aggregates			
	Slag	Fly ash	Nano-s	NaOH	H ₂ O		0–2	2–4	1–2	0.5–1
D15-5.0	384	96	170.8	30.9	77.7	370	200	150	30	
D15-3.5	384	96	119.6	21.7	119.1	380	200	150	30	
D15-2.0	384	96	68.3	12.4	170.7	390	200	150	30	
D12-5.0	384	96	170.8	30.9	77.7	0	240	180	30	
D12-3.5	384	96	119.6	21.7	119.1	10	240	180	30	
D12-2.0	384	96	68.3	12.4	170.7	20	240	180	30	
D14-3.5	384	96	119.6	21.7	119.1	260	230	140	30	
D13-3.5	384	96	119.6	21.7	119.1	135	245	150	30	

using a heat transfer analyser (ISOMET 2104) [38]. The acoustic absorption coefficient was measured according to EN 10534-2 [39]. Microstructure of the reaction products was identified by scanning electron microscopy (SEM) using a JEOL JSM-IT100 instrument, operated with high vacuum mode at an accelerating voltage of 10 kV.

3. Results and discussion

3.1. Compressive strength

The 7 and 28 d compressive strength of mixtures with different density levels and a Na_2O content of 3.5% are shown in Fig. 1. The relations between the oven dry density and strength are briefly depicted. It can be seen that as the density decreases, there is an obvious reduction in strength at both 7 and 28 d. The highest 28 d strength reaches 30.7 MPa in mixtures with a density of $1471 \text{ kg}/\text{m}^3$, and it gradually reduces to 20.6 MPa in samples with a density of $1163 \text{ kg}/\text{m}^3$. The relation between density and strength in this study is in line with previous studies concerning the lightweight aggregate based composites [40–44], while the strength results shown here are higher in general than the alkali activated lightweight composites reported in literatures [45–47]. Considering the fact that those mixtures are having the same binder content, the reduced compressive strength is mainly attributed to the replacement of normal sand by lightweight aggregates (as shown in Table 2), and a less overall capacity of the aggregates against compressive loading is resulted. The 7 d strengths share a

similar tendency with the 28 d's, which decreases from 27.4 MPa to 18.5 MPa within the density range of around $1500\text{--}1200 \text{ kg}/\text{m}^3$. The 7 d compressive strengths shown in Fig. 1 are all above 88% of the 28 d strength. This is because of the nature of the alkali activated binder system and the ceiling effect of the lightweight aggregates, which presents a relatively fast reaction process and exhibits large percent of the strength at early ages [1,9,35]. It should be noticed that key parameters such as activator type and dosage, binder composition and fineness, curing conditions also strongly influence the reaction process, and therefore the strength development.

Fig. 2 depicts the effect of the equivalent Na_2O content on 28 d compressive strength of mixtures with two density levels: 1500 and $1200 \text{ kg}/\text{m}^3$, represented with sample label of D-15 and D-12 in the figure. As stated in the introduction part, the alkali activator contributes to a large fraction of the overall environmental issue of alkali activated materials, thus an efficient usage of the activator is of clear significance. For mixtures with a Na_2O content of 5% in this study, the additionally provided silicate from the activator accounts for around 14.9% of the total silicate within the system, and this activator dosage is commonly used in achieving a high strength [9,20,21,24–26]. When reducing the Na_2O content to 3.5% and 2.0%, the silicate fraction from the activator decreases to 10.9% and 6.6%, respectively. Concerning its influence on strength, as can be seen in Fig. 2 that there is an obvious increment of strength when increasing the Na_2O content from 2.0% to 3.5%, and the increase of strength exhibits a limited scale when further increasing the Na_2O content to 5.0%. To specify, in mixtures with

Fig. 1. Compressive strength of AA slag-fly ash lightweight composites with different densities.

Fig. 2. Compressive strength of AA slag-fly ash lightweight composites with different Na_2O contents.

a density level of 1500 kg/m^3 , the 28 d strength is increased from 24.0 MPa to 30.7 MPa when shifting the Na_2O dosage from 2.0% to 3.5%, namely a relative increase of 27.9%, and this value is 32.5 MPa in mixtures with a Na_2O content of 5.0%, with a relative increase of only 5.9%. This phenomenon reveals that both Na_2O content and density show an important influence on strength. On the one hand, increasing the alkalinity (Na_2O %) will promote the activation of the binder that consequently leads to a higher strength from the aspect of the binder matrix; while on the other hand, the usage of lightweight aggregate limits the strength development by the relatively low crushing strength of the aggregate. Investigating the effect of both density and alkalinity provides insights concerning how the strength would vary within the designed parameter ranges, which also gives information on tailoring the strength with certain densities and reasonable activator dosages for different application requirements.

3.2. Reaction kinetics

The isothermal calorimeter test was performed on mixtures with the Na_2O content of 2.0%, 3.5% and 5.0%, respectively, and lightweight aggregates were added with an aggregate/binder ratio of 0.8 (based on the mixture proportions shown in Table 2), in order to evaluate their effect on the early age reaction. Fig. 3 illustrates the normalized heat flows of samples with and without lightweight aggregates during the first 6 d; the heat flow is normalized by mass of the binder (slag and fly ash). The sample labelled as “LWA-5.0” refer to the one with Na_2O content of 5.0, and “LWA-5.0 + Agg” means lightweight aggregates are included in the test. It is generally agreed that reaction process of alkali activated materials can be classified into four typical stages [18,48,49]: the initial dissolution period, induction period, acceleration/deceleration period and stable period. The first stage refers to the initial contact of the solid components with liquid solution, and the dissolution of the precursors. The induction period refers to the temporary induction of heat release before the main reaction takes place. The acceleration and deceleration periods are attributed to the massive formation of the reaction products, and the stable period indicates that the main reaction process comes to the end while minor reaction may still occur in the long term.

It can be seen from Fig. 3 that the mixtures with a Na_2O content of 5% exhibit a main reaction peak at around 16 h, with a peak intensity of 1.35 mW/g . The induction stage shows an induction

period between around 6 and 10 h, followed by an evident increase of heat flow representing the intensive chemical reaction [50]. When reducing the Na_2O content to 3.5%, a significantly retarded reaction process can be observed. The induction period appears at around 9–22 h, the beginning of this stage is 3 h later than the one in sample with a Na_2O content of 5%, also the duration of this stage is more than 3 times longer. The location of the main reaction peak is also delayed to 37 h, which is 21 h later than the 5% Na_2O mixtures. Besides, the peak intensity is reduced to 0.63 mW/g and the deceleration period exhibits a more moderate reduction of heat release. A further lowered Na_2O dosage of 2.0% results in a dramatic delay of the reaction process. For instance, the induction period lasts more than 48 h; the main reaction peak exhibits an obviously broader covered area with a low peak intensity of about 0.34 mW/g , indicating a gradual and slow formation of the reaction products. Until the testing age of 144 h, the deceleration stage is still ongoing. Thus it can be concluded that the reduction of the reaction process does not present a linear relation with the Na_2O content, the shift of Na_2O concentration effectively influences the characters of the reaction process such as induction time, reaction intensity, the location and duration of main reaction period. Similar tendencies are also shown in previous researches regarding the effect of activator modulus (usually with a constant Na_2O dosage) [35,49], while the reduction of peak intensity and the delay of the main reaction process is rather limited compared to the case of this study, which suggests that the activator characters such as additional silicate content and the alkalinity strongly control the activation process of the precursors.

The reaction kinetics results can be correlated to the compressive strength results in this study, where mixtures with a Na_2O content of 2.0% show an obviously lower strength than the ones with higher Na_2O contents. It is concluded that the delayed reaction process together with lower intensities provide a reduced activation on the starting materials, resulting in an overall lower reaction degree and strength. The heat release curves with dash lines refer to the mixtures with lightweight aggregate additions. For mixes with a Na_2O content of 5.0%, the aggregate incorporation slightly reduces the intensity of the main reaction peak without significant effect on its location, and there is also a very slight delay of the induction period. Similar trends are also shown in samples with 3.5% Na_2O content, indicating that the effect of lightweight aggregate on the early age reaction is rather limited, and those slight effects are probably attributed to the absorption of small amount of activator during the initial mixing. Mixes with a Na_2O content of 2.0% exhibits an evident delay of the main reaction peak, which indicates that under low alkalinity conditions, the effect of lightweight aggregate (activator absorption) is magnified.

The cumulative heat release of all mixtures is shown in Fig. 4. Differences in heat release processes caused by Na_2O content and aggregate addition are clearly depicted. Mixtures with a Na_2O content of 5.0% present the highest cumulative heat of about 166 J/g till the testing age of 144 h. Samples with 3.5% Na_2O show a slightly lower heat release of 153 J/g , while the heat release of mixes with a Na_2O dosage of 2.0% is significantly lower: 106 J/g , which is also partly because that the main reaction process is still processing. When lightweight aggregate is incorporated, similar heat release is shown in 5% Na_2O mixtures. Similar to the results shown in Fig. 3, as the Na_2O content decreases, the effect of lightweight aggregate on heat release becomes more significant.

3.3. Gel structures

In order to investigate to the effect of the Na_2O content on the gel compositions, the TG/DTG and FTIR analyses were performed and the results are shown in Figs. 5 and 6, respectively. Samples with the label of “LWA-2.0” refer to the ones produced with

Fig. 3. Normalized heat flows of AA slag-fly ash pastes with different Na_2O contents and lightweight aggregates.

Fig. 4. Normalized heat releases of AA slag-fly ash mortars with different Na_2O contents and lightweight aggregates.

Na_2O content of 2.0%. As shown in Fig. 5(a), the mixtures with 3 levels of Na_2O content exhibit an evident mass loss before around 100 °C, owing to the evaporation of physically bound water. Another significant loss in mass is shown between about 250 and 450 °C, which are attributed to the decomposition of the reaction products. Afterwards, a gradual decrease in mass is presented in general until 1000 °C; while the remarkable mass loss between 600 and 800 °C is assigned to the decomposition of carbonates [51], indicating that a certain degree of carbonation took place during the curing process. Three mixtures exhibit similar mass loss before 100 °C (mainly evaporable water), which is 4.9%, 4.0% and 5.3% for samples with a Na_2O content of 5.0%, 3.5% and 2.0%, respectively. This value can be slightly influenced by sampling and variations, thus it is difficult to precisely identify the effect of Na_2O dosage in this mass loss range. The mass loss between 100 and 600 °C, which can be mainly attributed by the decomposition of reaction products, is 8.0% for mixes with a Na_2O content of 5.0%. It slightly decreases to 7.4% in mixes of 3.5% Na_2O ; while this

value is 5.4% in the case of 2.0% Na_2O , showing a significant reduction. It indicates that besides the delayed reaction process within the first 144 h, the amount of reaction products in 2.0% Na_2O mixes after 28 d of curing is also obviously lower. The mass loss between 600 °C and 1000 °C is partly attributed to the decomposition of reaction products, while the carbonates also play a role. A detailed calculation in the temperature range of 600 °C and 800 °C shows that when increasing the Na_2O dosage from 2.0% to 5.0%, the mass loss in this range is increased from 0.5% to 1.0% and then 1.4%, which could be an indication that high alkaline condition contributes to the carbonation of alkali activated materials. The DTG curves shown in Fig. 5(b) present three evident peaks, the one that located around 100 °C refers to the obvious mass loss of bound water. The shift of this peak to higher temperatures indicates a gel structure with more tightly bound water and finer pore structures [52,53]. The peaks shown between 250 and 450 °C verify the decomposition of certain reaction products, which are reported to be the dehydration of hydrotalcite that is typically formed from alkali activated slag [54]. The intensity of the peaks around 600 °C and 800 °C indicates deferent degrees of carbonation caused by the Na_2O dosage.

The infrared spectra of the unreacted slag and fly ash, and the reaction products with different Na_2O contents are shown in Fig. 6. The original slag presents a broad main vibration band at around 900 cm^{-1} , which is attributed to the vibration of terminal Si-O bonds [55]. Another obvious peak at around 670 cm^{-1} is assigned to the stretching vibration of T-O groups [56]. The unreacted fly ash exhibits several shoulders at around 1080 cm^{-1} and between 600 and 800 cm^{-1} , which can be attributed to the presence of crystalline phases such as quartz and mullite [57]. The main absorption band at around 1020 cm^{-1} is assigned to the Si-O-T bonds with bridge conditions [58]. Upon activation, all mixtures present similar typical absorption bands regardless of the Na_2O contents. The slight absorption band at 1420 cm^{-1} confirms the presence of carbonates that are observed from the TG analysis, and the band at 1640 cm^{-1} indicate the presence of water. All mixes show a main absorption band around 950 cm^{-1} , which is the vibration of a non-bridging Si-O bond [59], also commonly recognized as C-A-S-H type gels. The fixed location of the typical bands together with the TG-DTG results indicate that the Na_2O

(a)

(b)

Fig. 5. TG-DTG analysis of AA slag-fly ash blends with different Na_2O contents.

Fig. 6. FTIR spectra of the starting materials and reaction products.

content affects the reaction degree and carbonation behaviours without changing the typical silicate structures within the reaction products.

3.4. Thermal conductivity

In terms of the building materials, a low thermal conductivity contributes to an enhanced indoor thermal comfort, saving the energy cost and preventing the fire caused collapses; while lightweight concrete products based on alkali activated materials are capable of achieving those requirements with a further lowered environmental impact. Fig. 7 shows the thermal conductivity of samples with a Na_2O content of 3.5% and different density levels used in this study, and a comparison with the available literature [60–63]. A reduction of thermal conductivity with the decrease of the density can be observed in general. For mixtures with a density level of round 1500 kg/m^3 , the thermal conductivity (k -value) is $0.37 \text{ W/(m}\cdot\text{k)}$, this value is lower than the ones from the obtained literatures. This is because that besides the density, the differences in matrix composition, type of binder and aggregates also show an influence on the property of thermal insulation [64,65]. The ther-

Fig. 7. Thermal conductivity of mixtures with different densities.

mal conductivity reduces to $0.29 \text{ W/(m}\cdot\text{k)}$ in samples with a density level of 1400 kg/m^3 , with a reduction rate of 21.6%. As can be seen from the mix proportions shown in Table 2, once the Na_2O content is fixed, the difference between different mixes lies in the aggregate type and dosage. The reduction of density from around 1500 to 1400 kg/m^3 results in a decreased standard sand dosage of 31.6% by mass, indicating its negative influence on thermal insulation. Further reduction of the density to around 1300 and 1200 kg/m^3 leads to further reduced thermal conductivities of 0.22 and $0.16 \text{ W/(m}\cdot\text{k)}$, respectively. It should be noticed that the thermal conductivity of $0.2 \text{ W/(m}\cdot\text{k)}$ can be classified to the T2 level of thermal insulation products based on the standard of EN 998-1 [66]. It can be seen from Fig. 7 that the thermal conductivity values from this study are lower than that from the literature, which may indicate the advantage of using the alternative binder system together with the lightweight aggregate in this case. Overall, the compressive strengths of around 20–30 MPa together with moderate densities and ideal thermal conductivities indicate a wide and promising application potential of this new lightweight concrete.

3.5. Acoustical absorption

Owing to the massive addition of the porous lightweight aggregates, the resulting alkali activated lightweight concrete is expected to exhibit good sound absorption behaviours. Fig. 8 exhibits the sound absorption coefficient as a function of frequency, four mixtures with a Na_2O content of 3.5% with different density levels were tested. For instance, the mixture with label of D-15 refers to the sample with a density around 1500 kg/m^3 . Generally speaking, the absorption coefficient is significantly increased with the decrease of density, in another word with the increase of lightweight aggregate content within the matrix; and all mixtures mainly show absorption of sound with medium frequencies between around 300 and 1700 Hz. Specifically, mixtures with a density of round 1500 kg/m^3 present a peak absorption coefficient of 0.2, between 800 and 1000 Hz. The peak absorption coefficient increases to around 0.35 and 0.52 in samples with a density about 1400 and 1300 kg/m^3 respectively, while the main absorption frequency range remains similar. A further reduction of density to around 1200 kg/m^3 results in an increased absorption coefficient to above 0.7, with shifted main absorption frequency to higher values. In terms of the low frequency and high frequency ranges, this lightweight product shows limited sound absorption effect. It

Fig. 8. Acoustic absorption of mixtures with different densities.

should be mentioned that the medium frequency usually refers to the sound from humans and daily life.

3.6. SEM analysis

Scanning electron microscopy images are used to characterize the applied lightweight aggregate and the interfacial transition zones (ITZ) of the reaction products. Due to the fact that the effect of Na_2O content on micro scale characteristics is not significant, the

reaction products shown in Fig. 9 are having a constant Na_2O content of 3.5%. Fig. 9-A presents the micro graph of the lightweight aggregate surface, porous structures with irregular shaped surfaces can be observed. This porous structure may already result in a direct link of the aggregate surface and the internal areas. Fig. 9-B depicts a sectional view of the internal structure of the lightweight aggregate, micro pores with different sizes and shapes are clearly presented. It is clear that this lightweight aggregate exhibits a high porosity, which would result in excellent thermal insulation

Fig. 9. SEM images of lightweight aggregates and the transition zones.

Table 3Calculation on the carbon footprint (kg/m^3).

Sample	Precursors	Activator (exclude H_2O)			CO ₂ emissions from activator	CO ₂ emissions from binder	Total CO ₂ emissions
		Olivinenano-s	Water glass	NaOH			
D15-5.0-NS	384 Slag + 96 Fly ash	32.5	0	30.9	74.2	57.5	131.7
D15-5.0-WG		0	42.3	18.3	99.1		156.6
D15-3.5-NS		22.7	0	21.7	52.0		109.5
D15-3.5-WG		0	29.6	12.8	69.3		126.8
D15-2.0-NS		13.0	0	12.4	29.7		87.2
D15-2.0-WG		0	16.9	7.3	39.6		97.1
Ref-OPC	480 OPC	0	0	0	0	393.6	393.6

and sound absorption properties in macro scale. This sectional view also demonstrates that the micro pores within the aggregate are more like single-direction distributed tube shaped structures, rather than individual spherical ones. The ITZ shown in Fig. 9-C verifies the existence of tube shaped pore structures, as can be seen that two lightweight aggregates that are separated by the binder matrix exhibit different morphology. Together with Fig. 9-D it can be observed that the boundary between aggregate and the pastes is not clearly defined. Fig. 9-E shows that certain amount of reaction products is grown within the porous structures that are close to the ITZ. However it is difficult to identify its effect on the mechanical properties, since on the one hand the reaction products break the original structure of aggregate, while on the other hand the formed reacted gels at those areas strengthen the ITZ by densifying the original porous surface of the aggregates. Lightweight aggregates with smaller particle sizes experience a more evident interaction with the binder matrix, as can be seen from Fig. 9-F that the porous structure of smaller sized lightweight aggregates are partly (sometimes fully) occupied with the reacted gels. This may lead to an increment of the density compared to the designed one, and may slightly reduce the thermal insulation and sound absorption properties.

3.7. Advantages in carbon footprint

Applying alkali activated materials shows an advantage in carbon emission towards Portland cement. While within the alkali binder systems, the Na_2O content is directly linked to the environmental issues. Moreover, the replacement of olivine nano-silica to commercial waterglass will further reduce the overall carbon emission. A calculation on the CO₂ footprint of the mixtures used in this study is shown in Table 3. The carbon footprint is usually calculated as the total CO₂ emissions in unit volume of concrete. The assumed recipes are based on the mix proportions shown in Table 2. Sample with the label of “D15-5.0-NS” refers to the one with density level of $1500 \text{ kg}/\text{m}^3$, Na_2O content of 5.0% and activator type of nano-silica, and “WG” refers to the activator of “water glass”. The carbon footprint used in this calculation is $1.915 \text{ tCO}_2/\text{t}$ for NaOH, $1.514 \text{ tCO}_2/\text{t}$ for commercial waterglass, $0.82 \text{ tCO}_2/\text{t}$ for Portland cement, $0.143 \text{ tCO}_2/\text{t}$ for slag and $0.027 \text{ tCO}_2/\text{t}$ for fly ash [67,68]. Concerning the olivine nano-silica, a CO₂ footprint of $0.461 \text{ tCO}_2/\text{t}$ is taken from the LCA analysis in VTT (EU F7th project, ProMine internal report) [69], based on an average European industrial case scenario. The values shown above are based on the effective solids contents excluding water.

It can be observed from Table 3 that the carbon emission of Portland cement is few times higher than alkali activated materials; the contribution of CO₂ is all from the cement clinkers. In terms of the alkali activated materials, the carbon emission from the binder accounts for around 37–69% of the overall emissions. When reducing the Na_2O contents from 5.0% to 2.0%, the carbon

emission from the commercial sodium silicate based activator can be reduced by 60%, such a big reduction of carbon footprint may bring attentions on using suitable Na_2O dosages and reaching a balance between performances and environmental impact. Moreover, when olivine nano-silica is applied as commercial waterglass replacement, the carbon emission in terms of activator can be further reduced by around 25%. In overall, those reduced carbon emissions from Na_2O content and alternative silica source provide great potentials for the future mix design and applications.

4. Conclusions

This paper evaluates the mechanical properties, thermal property, acoustical absorption and interfacial transition zones of eco-friendly alkali activated slag-fly ash lightweight composites (LWC) with different density classes. The effect of Na_2O contents and the utilization of alternative silica source on early age reaction, gel characteristics and carbon footprints on the designed LWC are assessed. Mixtures with 28 d compressive strength up to 32.5 MPa and densities between 1200 and $1500 \text{ kg}/\text{m}^3$ are resulted, and a direct correlation between strength and density is observed. The reduction of Na_2O content from 5% to 2% significantly prolongs the reaction process, reduces the content of bound water, hydrotalcite and carbonates, but shows negligible effect on the silicate structure. Compared to the commercial waterglass, the utilization of olivine nano-silica reduces the carbon footprint from activator by around 25%. The lightweight concretes exhibit very low thermal conductivity between 0.16 and $0.37 \text{ W}/(\text{m}\cdot\text{k})$, as well as good sound absorption coefficient up to 0.7 for medium frequencies. The application of alkali activated binders and applying this activator to lightweight concrete suggests an additional approach of preparing construction and building materials with multifunctional properties and greatly reduced environmental impacts.

Conflict of interest

None.

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