Lightweight Cement-Based Composites Incorporating Hollow Glass Microspheres: Fresh and Hardened State Properties

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ABSTRACT

This research aims to develop a lightweight cementitious composite with satisfying mechanical and good thermal insulating properties. Two different types of hollow glass microspheres (HGM) were used as lightweight aggregates and were substituted with fine aggregate by 10, 20, and 40% by volume. The rheological, physical, mechanical, and microstructural properties of the resulting HGM-incorporated composites were investigated and correlations were established between physical and mechanical test results. The results showed that the physical and mechanical properties of individual HGM particles play a dominant role in the properties of lightweight mortars. HGM addition provided reductions up to 20% in the density and 45% in the thermal conductivity values of mortars compared to the reference. The best HGM ratio in the tested range was found as 20%, which provides benefits such as reduced density and enhanced thermal insulation capability without causing a significant reduction in compressive strength. It was concluded that HGMs can be used in the lightweight cementitious mortar production which has great potential in building applications to reduce the heating energy consumption.

Keywords: Hollow glass microspheres, rheology, mechanical properties, thermal conductivity, microstructure.

1. INTRODUCTION

The advantages acquired by using lightweight materials in buildings are the reduced weight and the dead load acting on the structures which result in reduced size of structural elements [1], and lower thermal conductivity which accordingly increases energy savings and improves fire resistance. Reducing the greenhouse gas emissions related to energy consumed during operational issues of a building is crucial considering sustainability issues [2]. Therefore, any material related strategy that can help develop thermally efficient materials with reduced weight and that are structurally sound can also help with optimal building design from multiple performance points of view.

Lightweight concrete (LWC) is usually manufactured by adding lightweight aggregates (LWA) into the cementitious matrix. The most widely used LWA are pumice [3], expanded perlite [4–6], shale [7–9], expanded clay [10], and diatomite [11]. While the incorporation of lightweight materials in cementitious matrix achieves reduced density, traditional LWC and other cementitious composites possess lower mechanical properties. Recently, lightweight fillers (LWF) such as polystyrene beads, thermoplastic microspheres [12], hollow glass microspheres (HGMs) [13–15] and fly-ash [16–18] have been used to manufacture high performance lightweight cementitious composites for structural applications in buildings to compensate the disadvantages of conventional lightweight aggregate concrete.

The HGMs, also known as glass bubbles, can be defined as a glass material filled with air encapsulated by a thin spherical glass enclosure. HGMs are made from soda-lime borosilicate glass, and depending on their characteristic properties they possess different strength and density. They are nonporous, chemically inert, and resist against water and oil [19]. HGMs are commercially available with a variety of particle size and density and are compatible with cementitious materials [20].

HGMs are generally utilized as fillers [14] owing to their unique performance such as chemical, corrosion and fire resistance, light quality, and superior mechanical and physical properties [19,21,22]. Moreover, the spherical shape and the smooth surface of HGM has yielded to their use in oil well cement slurry [23]. It has been reported that HGM particles may cause an alkali-silica reaction in cement mortar [24], therefore before using HGMs in cementitious materials their chemical reactivity with cement compounds should be researched. In addition, the smooth surface, as well as poor bonding interface between matrix phase and HGM, may lead to lower strength and brittle failures [15].

The literature presents only a few studies on the use of HGMs in cementitious composites. Perfilov et al. [25] showed the potential use of HGM to cement oil and gas wells up to 4000 m deep. Al-Gemeel et al. [20] researched the effect of HGM on the mechanical and fracture properties of hybrid fiber reinforced engineered cementitious composite. They found reduced mechanical properties with increasing HGM content, but enhancements in mechanical properties, as well as reductions in density, could be provided with reducing the water to cement ratio. The authors concluded that it was possible to manufacture viable lightweight engineered cementitious composite products using HGM and suggested the need for further study on the feasibility of incorporating a higher amount of HGM in cementitious composites. Brooks et al. [2] performed extensive experimental research to comparatively study the effect of various types of LWF including the HGM on the several physical and mechanical properties of lightweight composites. They found that the thermal properties are significantly

affected by the volumetric ratio of the LWF and the mechanical properties are greatly dependant on the LWF particle properties including the bond between the particles and the cement matrix. Their research also showed that the HGM particles with lower shell thickness to diameter ratio tend to break when subjected to mechanical loads which should also be considered when using such materials in cementitious composites. Yun et al. [13] incorporated HGM at varying ratios up to about 30% by volume of total aggregates in lightweight aggregate concrete and found that HGM helped to reduce the thermal conductivity of concrete and they noted that to satisfy the mechanical soundness, the optimum HGM addition should be no more than 20%.

Limited information exists in the literature regarding the effect of LWA on the rheological properties of cementitious materials. Senff et al. [26] studied the effects of LWAs such as perlite and vermiculite on the rheological and mechanical properties of cement-based mortars. They reported that both types of aggregates improved the rheological properties substantially. On the other hand, the use of LWA degraded the mechanical resistance of mortars. Assaad [27] investigated the effect of styrene-butadiene rubber (SBR) latexes on the rheology of lightweight self-consolidating concrete mixes and found that SBR addition leads to reduced concrete flow rate and passing ability. The author attributed that to the increased cohesiveness resulting from the coalescence of water-soluble latexes and binding of the cementitious matrix. Gogoi et al. [28] researched the synergistic effect of HGM fillers and short bamboo fibers (SBF) on rheological and mechanical properties of light-weight composite materials. The authors found that HGM addition caused a significant improvement in hardness while a marginal decrease on impact strength was observed. They also concluded that both HGM and SBF improved the rheological properties.

The papers reviewed above focus on the potential use of LWAs, LWFs, and HGMs in cementitious materials and their effects on the rheological, mechanical, physical, and thermal properties of cementitious composites. However, the effects of HGM inclusion on the rheological, physical, and thermal properties of cement-based composites have not been revealed in detail yet. Considering the lack of studies on the rheological and thermal properties of HGM-modified cement composites, this experimental study aims to give a contribution to the improvement of thermal and physical properties of HGM-modified lightweight mortar without causing a significant reduction in the compressive strength. Therefore, the effect of two different types of HGMs (differing with their particle size, density, thermal conductivity, and crushing strength) on the fresh and hardened state properties of cement paste and mortar were investigated. Moreover, the best ratio for the HGM incorporation was proposed to give a satisfactory mechanical performance with reduced density and thermal conductivity. It was anticipated that the results obtained from this study will provide as yet unknown details on these lightweight cementitious systems and will contribute to the literature.

2. EXPERIMENTAL DETAILS

2.1. Materials and Mixtures

CEM I 42.5R type Portland cement was used as a binder throughout this study. The physical and chemical properties of cement are listed in Table 1.

CEN Standard sand with a maximum particle size of 2 mm and particle density of 2.68 kg/dm^3 was used as fine aggregate. The particle size distribution of HGMs (coded as HGM-I and HGM-II), cement, and sand were determined with Malvern Mastersizer 2000 instrument and the results are presented in Fig. 1. Fig. 2 shows the SEM micrographs of the HGMs used in this research. It can be seen that both HGMs have a spherical shape, HGM-I has finer diameter compared to HGM-II, and HGM-II presents a few amounts of damaged particles in its natural state.

Table 1 - Chemical composition and physical properties of cement

Compound (%)		
	SiO_2	18.9
	Al_2O_3	4.8
	Fe_2O_3	3.3
	MgO	1.5
	SO_3	3.3
	Na_2O	0.4
	CaO	63.7
	K_2O	0.7
	TiO_2	-
	Mn_2O_3	-
	S	-
	Cl (ppm)	600
LOI (%)		2.9
Specific surface area (cm^2/g)		3854
Specific gravity		3.1
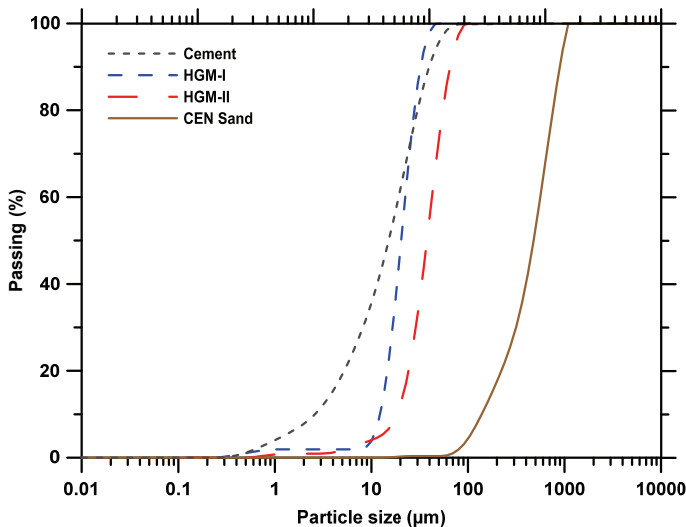
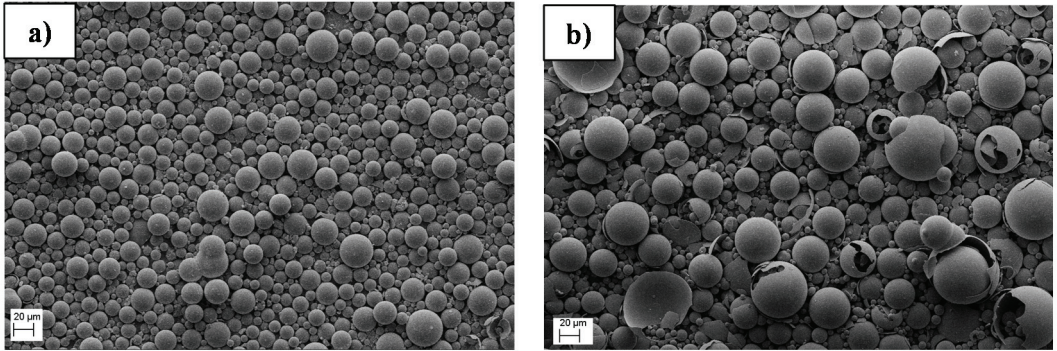*Fig. 1 - Particle size distribution of solid materials*

Fig. 2 - SEM images of (a) HGM-I and (b) HGM-II

The physical and mechanical properties of HGMs obtained from the manufacturer are summarized in Table 2. HGM-I can be distinguished from HGM-II with its finer particle size, higher isostatic crush strength, higher density, and higher thermal conductivity coefficient. The median particle size (d_{50}) of sand, cement, HGM-I, and HGM-II was determined by laser diffraction as 479.0 μm, 15.1μm, 20.3 μm and 37.5 μm, respectively.

Table 2 - Physical and mechanical properties of HGMs

HGM code	Isostatic crush strength (MPa)	Density (g/cm^3)	Specific surface area (cm^2/g)	Thermal conductivity (W/mK)
HGM-I	110.3	0.46	5630	0.153
HGM-II	2.8	0.22	2730	0.076

HGMs were introduced into the mortars substituting the sand by 10, 20, and 40% by volume by following a similar methodology from previous studies to maintain the compressive strength [2, 25]. It is well known that the cement replacement method might cause high reductions in strength [30] and therefore aggregate replacement method might be a better way of utilizing inert inclusions in cement composites to avoid significant reductions in mechanical properties [31–33].

A total of seven mortar and paste mixes were obtained and coded as Ref (reference mortar without HGM), HGM-I-10, HGM-I-20, HGM-I-40, HGM-II-10, HGM-II-20 and HGM-II-40, where the numbers represent the volume ratio of HGM. The mortar samples were prepared with a constant paste and aggregate volume of 50% and the HGMs replaced the fine aggregate at three different volumetric ratios as mentioned before. The water to cement ratio was considered as 0.5 in all mixtures.

The mixing started with a dry mix of solid materials (cement, sand, and HGM) for about one minute, then the water was added and the mixing was continued for about another three minutes until a uniform mix was obtained. Paste samples incorporating HGM were also manufactured to analyse the rheological properties and the microstructure. The paste samples were stored in sealed conditions at room temperature until being tested at 28 days for the microstructural analysis. Mix proportions of mortar and paste mixtures are shown in Table 3. The alkali-silica reactivity of the HGMs used in the experimental study was researched as per ASTM C1260 before starting the tests and the volume expansion was found to be below 0.09%.

Table 3 - Mixture proportions (kg) of mortar and paste samples

| Mixture code | Cement | | Water | | Sand | | HGM | | | |
| | | | | | | | Mortar | | Paste | |
	Mortar	Paste	Mortar	Paste	Mortar	Paste	HGM-I	HGM-II	HGM-I	HGM-II
Ref	607.8	1215.7	303.9	607.8	1340	0	0	0	0	0
HGM-I-10	607.8	1215.7	303.9	607.8	1206	0	23	0	41.8	0
HGM-I-20	607.8	1215.7	303.9	607.8	1072	0	46	0	76.7	0
HGM-I-40	607.8	1215.7	303.9	607.8	804	0	92	0	131.4	0
HGM-II-10	607.8	1215.7	303.9	607.8	1206	0	0	11	0	20
HGM-II-20	607.8	1215.7	303.9	607.8	1072	0	0	22	0	36.7
HGM-II-40	607.8	1215.7	303.9	607.8	804	0	0	44	0	62.9

2.2. Experimental Procedures

The effect of HGM inclusion on the rheological properties of cement paste was determined at room temperature (20 ± 0.5 °C) by using a rheometer with a strain-controlled mechanism. A four-blade vane configuration was used in this study. Approximately 35 ml of paste was introduced into the rheometer cup and precautions were taken to do the measurements immediately after the mixing. The test procedure consisted of three main stages; (*i*) constant pre-shear at 100/s for 30 s to homogenize the paste, (*ii*) 30 s resting, followed by a (*iii*) stepped ramp up from 0.05-to-100/s, and a stepped ramp down from 100-to-0.05/s (Fig. 3). At each step, steady-state shear stress was achieved after 10 sec. Each shear rate was maintained for 20 sec, data were acquired every second and the average of last 10 consecutive data was used to extract the yield stress (YS) and plastic viscosity (PV) in the down ramp. To obtain a reasonable portion of the stress plateau, a wide range of shear rate was applied as generally considered in the ordinary Portland cement pastes as well [34–40].

The plateau region of the ramp down was considered to be the YS in logarithmical mode. PV was calculated as the slope of the linear proportion of shear rate between 25/s and 100/s.

The workability of mortars was assessed by flow test as per EN 1015-3. Fresh mortar samples were placed into a conical mould with bottom and top diameter of 100 mm and 70 mm, respectively, and with a height of 60 mm. The excess mortar at the top of the cone was

removed and the mould was raised. The flow diameter was measured in two directions to determine the average flow diameter of the mortar. The compressive strength of mortar samples was determined on 50 mm cube specimens as per ASTM C109 at 3, 7, and 28 days and the flexural strength was determined at 28 days on $40 \times 40 \times 160$ mm prismatic specimens as per EN 196-1.
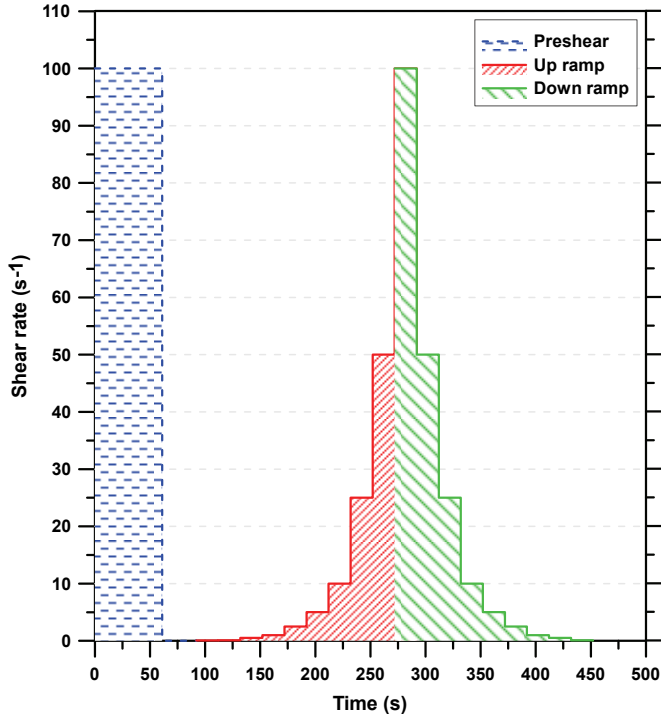

Fig. 3 - Rheological test procedure

The thermal conductivity measurements were performed on oven-dried disc specimens with a nominal diameter of 50 mm and thickness of 25 mm using the *Lasercomp Fox* series heat flowmeter instrument following the ASTM C518, EN 12664, EN 12667 and ISO 8301 standards. The thermal conductivity was determined by the following Eq. (2.1):

$$q = -kA\frac{\Delta T}{\Delta x} \tag{2.1}$$

where *q (W/m²)* is the heat flow, *k (W/mK)* is the thermal conductivity, *A (m²)* is the area, *ΔT (K)* is the temperature difference and *Δx (m)* is the sample thickness.

The microstructure of the pastes incorporating HGM was analysed using a scanning electron microscope (SEM). The paste samples were crushed and small pieces taken from the core were used in the SEM analysis to illustrate the distribution of HGMs in the matrix and to analyse the microstructure.

3. RESULTS AND DISCUSSION

3.1. Rheological Properties and Workability

Rheological properties such as YS and PV were determined on fresh paste samples and the workability was determined on fresh mortar samples by flow test. Figs. 4-6 show the variation of the rheological properties and the flow diameter of the paste and the mortar samples according to the ratios of HGM.

Fig. 4 presents the typical flow curves of the HGM modified cement pastes. Shear-thinning behaviour was observed in all mixes. It can be seen that both HGM-I and HGM-II mixes consistently showed higher shear stress values at each shear rate compared to the reference mixes.
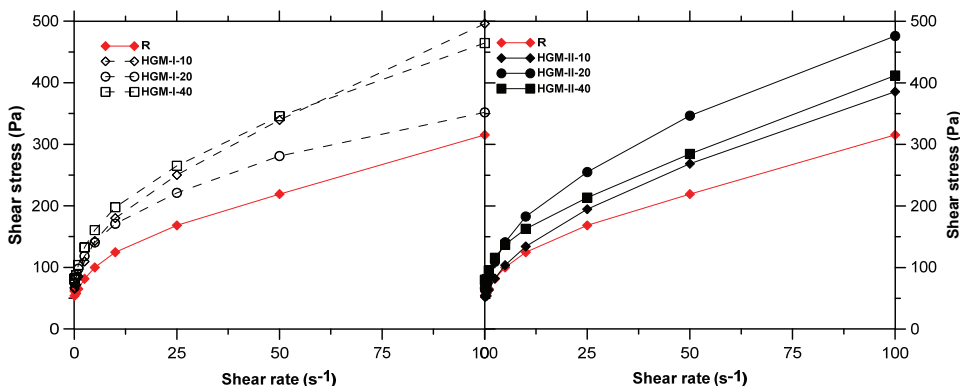

Fig. 4 - Typical flow curve of mixes

The YS values varied between 48.6 - 95.2 Pa and 44.3 - 63.3 Pa for HGM-I and HGM-II mixes, respectively. The YS and PV of the pastes consistently increased with increasing the HGM ratio; the reference mix (Ref) had the minimum YS and PV values, while HGM-I-40 and HGM-II-40 had the maximum values (Fig. 5). In addition, HGM-I mixes presented higher YS values than the HGM-II mixes, possibly due to the several characteristics of HGMs explained below. The PV results followed a similar trend as YS. HGM-I-40 mixes had the maximum PV value of 3.9 Pa.s, while the PV of the reference mix was only 0.3 Pa.s. The inclusion of HGM into the cement paste resulted in an increase in PV and this increase was significant when the HGM ratio increased from 20% to 40%. The increase in PV, with the increase in HGM volume from 20% to 40%, was 215% and 89% for HGM-I and HGM-II mixes, respectively.

It is generally known that an increase in particle concentration results in higher shear viscosity. One of the main reasons for the increase in YS and PV can be explained with the inter-particle forces between the HGM particles and the cement and HGMs. Inter-particle forces increase with the higher surface area of the components [41], therefore higher PV and YS can be expected in HGM-I mixes compared to HGM-II since HGM-I has a higher specific surface area (Table 2). Bentz et al. [42] reported that the rheological response is significantly influenced by both the particle densities and particle surface areas. Particle size distribution

(PSD) of the components of the blended mixes might be another parameter affecting the rheology. Lee et al. [43] noted that as the PSD of a cement/fly ash paste becomes wider, paste fluidity increases, which in turn reduces the apparent viscosity. Similarly, in this study, the PSD curves become narrower with HGM inclusion and consequently increases PV and YS. Since the YS values of the pastes were altered in the presence of HGM and the volume of the HGM was the same in HGM-I and HGM-II mixes, it can be concluded that the YS was mainly controlled by the fineness of the HGMs. Considering both the YS and the PV values of the mixes, it can be deduced that the rheological properties are greatly affected by HGM-I type HGM inclusion.
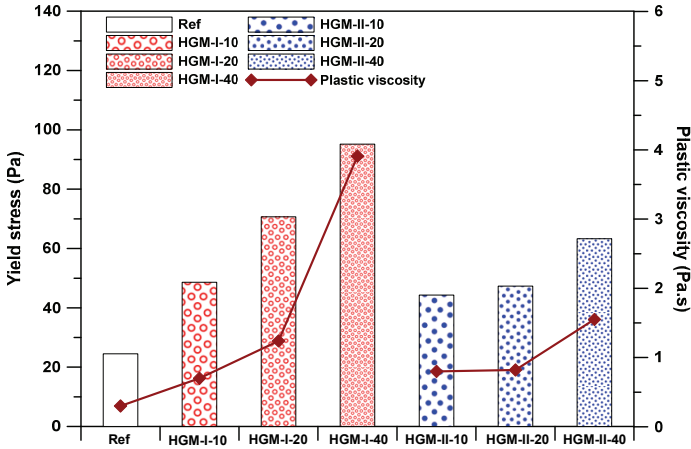

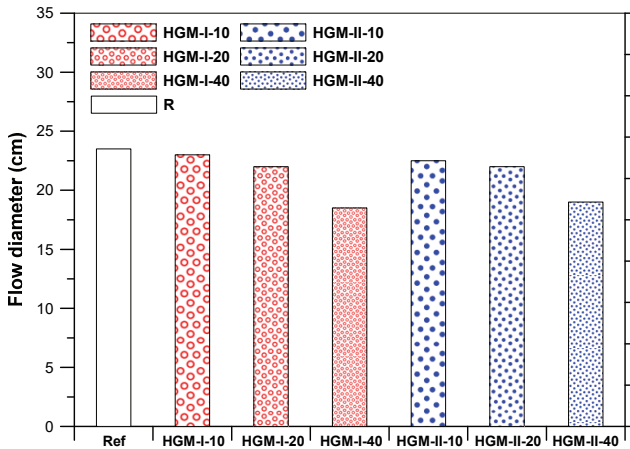*Fig. 5 - Yield stress and plastic viscosity of paste samples*

Fig. 6 - Flow diameter of mortar mixes

The workability test results are presented in Fig. 6. The flow diameter of mortars incorporating HGM reduced due to HGM inclusion. It could be thought that the incorporation of HGM may contribute to the flowability owing to its spherical shape. However, a significant reduction of about 20% (compared with the reference mortar) in flow diameter was noticed at 40% HGM ratio in HGM-I-40 and HGM-II-40 mixes. The reason for this behaviour was attributed to the fact that high amount of very small particles of HGMs negatively affects the workability and increases the water demand of mortar. On the other hand, lower HGM inclusion caused only slight reductions in workability (Fig.6).

Flow test results can be used to predict the rheological properties, such as YS, of the fresh cementitious materials [44–47]. Therefore, the results obtained in this research were used to establish a correlation between the rheological properties and flow diameter of HGM incorporated cement paste and mortars. The relationship between the determined rheological properties is presented in Fig. 7, where it can be noticed that they are correlated with each other, i.e. the higher the PV yields higher YS, and the higher the YS results in a lower flow diameter.
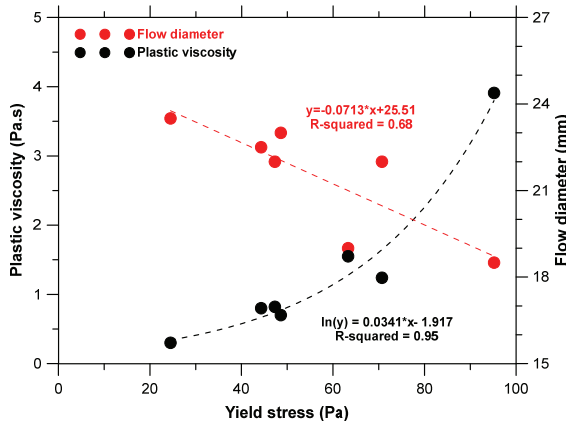

Fig. 7 - Correlations between plastic viscosity - yield stress and flow diameter - yield stress

3.2. Physical Properties

The physical properties of mortars are plotted in Figs. 8 and 9. Oven-dry density and thermal conductivity of the mortars proportionally reduced with the incorporation of both HGMs due to their low density and thermal conductivity. The reduction in the oven-dry density was relatively small for the ratios of 10 and 20%, but substantial reductions were recorded at higher ratios. The highest reduction (about 15% in HGM-I-40 and 20% in HGM-II-40) in the oven-dry density was observed at 40% incorporation ratio of both HGMs compared to the reference. The reduction in the thermal conductivity was found to be higher and the thermal conductivity consistently reduced by increasing the ratio of both HGMs. Thermal conductivity reduced by about 20% even at the lowest substitution ratio for each HGM, and the highest reduction was observed in the HGM-II-40 mix as 45%, compared to the reference. Comparing the effect of two HGMs on the thermal conductivity of mortars, it can be noticed

that the HGM-II mixes consistently possessed a lower thermal conductivity coefficient than the HGM-I mixes. The improvement in thermal conductivity is attributed to the lower thermal conductivity coefficient of the HGM-II particles.
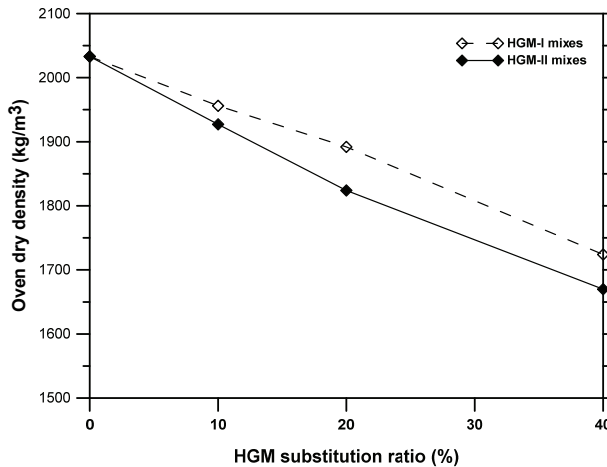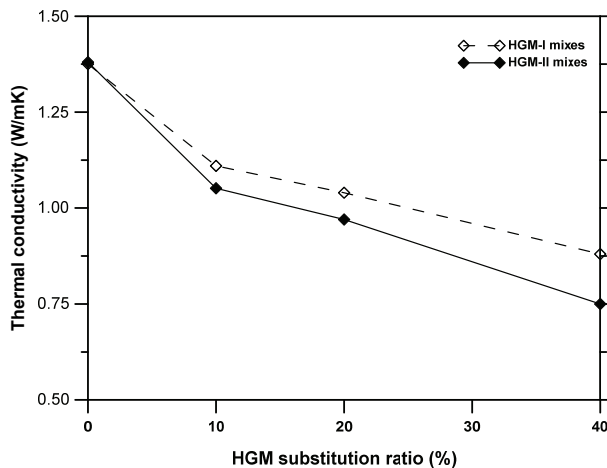

Fig. 8 - Oven dry density as a function of HGM ratio

Fig. 9 - Thermal conductivity as a function of HGM ratio

3.3. Mechanical Properties

Compressive strength development of the mortars as a function of age is presented in Figs. 10 and 11, where a consistent increase in compressive strength of mortars by hydration time was noted. HGM inclusion created significant strength reductions at 40% ratio. The reduction was 25% in HGM-II-40 and 16% in HGM-I-40 mixtures at 28 days compared to the reference (Table 4), nevertheless, the lowest compressive strength at 28 days was 43 MPa (HGM-II-40 mix). The literature presents similar reductions in compressive strength with HGM

incorporation. Al-Gemeel et al. [20] studied the effect of HGM addition on fiber-reinforced cementitious composites. Their research showed reduced compressive strength which was attributed to the increased voids in the cementitious matrix due to the hollow sphere structure of HGM. Zhang et al. [48] reported strength reductions when using excessive amounts of HGM in geopolymer matrix, but higher strengths with lower HGM dosage. Brooks et al. [2] analyzed the effect of several LWAs including HGMs on the physical and mechanical properties of mortars. Their research showed that the compressive strength of mortars depends highly on the particle size and the shell property of the LWA and that the mortars having smaller size HGM and thicker shells yield higher compressive strength. In addition, it is also reported that the smooth surface and poor bonding between HGMs and the cement matrix might be another reason for lower strength performance [15].
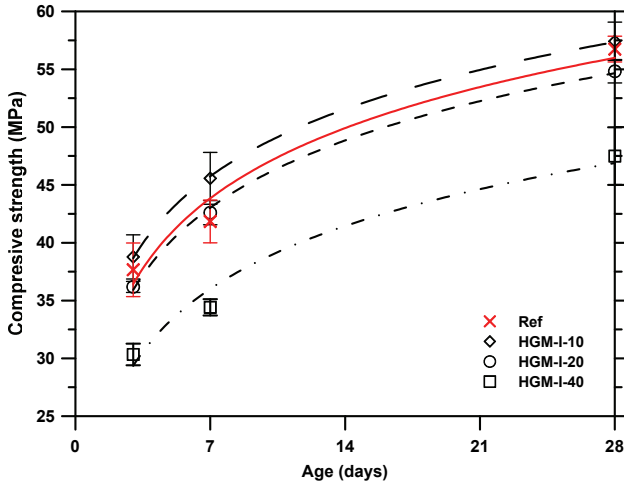

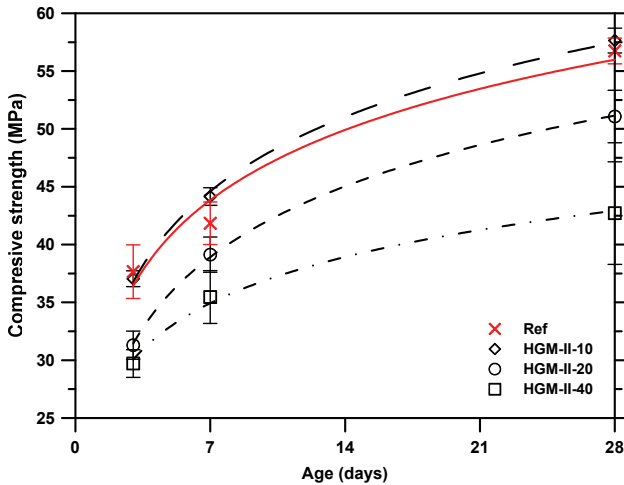*Fig. 10 - Compressive strength values of HGM-I mixes*

Fig. 11 - Compressive strength values of HGM-II mixes

The results of the present study show that at lower ratios (HGM-I-10 and HGM-II-10 mixtures) the compressive strength of mortars slightly increases compared to the reference (Table 4). The reduction in compressive strength was noted to be 3-10% at 28 days when the HGM ratio was 20%, which suggests a maximum substitution ratio with sand without sacrificing the compressive strength of cement mortar. It is also noteworthy to mention that the inclusion of HGM also reduces the density and the thermal conductivity of the mortar which also shows the probability of designing lightweight cementitious materials with lower density and higher thermal insulation properties without significant reduction in compressive strength. The effect of HGM type on the compressive strength results clearly shows that the mixtures incorporating HGM-I yields higher compressive strength compared to HGM-II. The main reason for this is possibly due to the higher strength property of HGM-I (about 40 times of HGM-II) particles and also its finer size distribution. It has been reported that the stress distribution of cementitious materials is related to both the sizes of inclusions and the difference in Young modulus of matrix and the inclusions and that the compressive strength increases with a decrease in LWA size [49]. Therefore, the finer particle size of the HGMs compared to the sand they substitute might have modified the stress distribution under compressive loads and improved the strength at especially lower substitution ratios. However, at higher ratios, the mechanical properties of HGM particles govern the compressive strength of the cement composites, and the achieved strength values of HGM incorporated mixes fail to surpass the reference mix where the fine aggregate is only sand.

Table 4 - Relative compressive strength of mortars

Mixture	Relative compressive strength (%)		
	3 days	7 days	28 days
Ref	100	100	100
HGM-I-10	103	109	101
HGM-I-20	96	102	97
HGM-I-40	81	82	84
HGM-II-10	98	106	102
HGM-II-20	83	94	90
HGM-II-40	79	85	75

The flexural strength results are shown in Fig. 12. A similar trend as in compressive strength was observed in flexural strength results of mortars. The flexural strength reduced with the HGM substitution, the exception was HGM-I-10, which showed a slight increase (about 3%) at 10% substitution ratio. The reduction in the flexural strength was noted as 13% and 31% for HGM-I-20 and HGM-II-20 mixes, and as 38% and 50% for HGM-I-40 and HGM-II-40 mixes respectively, compared to the reference. The higher strength loss in flexure compared to compression may indicate the weak interfacial bonding between HGM and matrix. The flexural strength of HGM-I mixes was found to be higher than that of the HGM-II mixes as in compressive strength at all HGM substitution ratios, which again indicates that the strength

characteristics of individual HGM particles play an important role in the mechanical properties of the cement mortars.
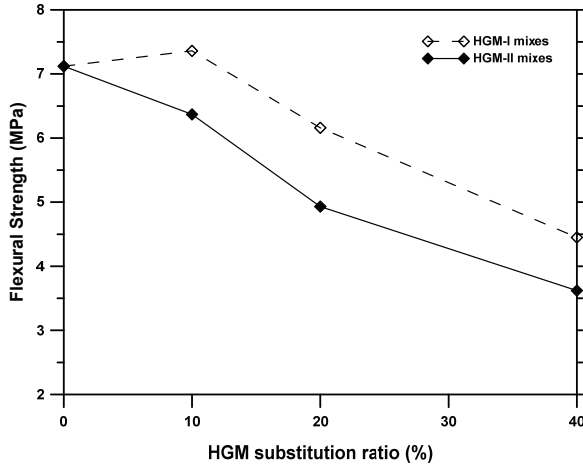

Fig. 12 - Flexural strength of mixes as a function of HGM substitution ratio

The most important function of incorporating LWA in the cementitious matrix is the reduction in the thermal conductivity coefficient with the reduced density. This beneficial reduction in the physical properties may unfortunately cause undesired reductions in mechanical properties, i.e. compressive strength, with the incorporation of LWA with weak strength properties. Table 5 summarizes previously researched different types of LWA in producing LWC for comparison with the HGMs used in this study. The general evaluation of the data presented in Table 5 shows that the compressive strength, density, and thermal conductivity reduces with an increase in the LWA content.

The strength reduction limits the use of these materials in structural applications where the compressive strength limit is set as 17-20 MPa in FIB and ACI codes [50]. Therefore the balance between strength, density, and thermal conductivity should be optimized with the aim of increasing strength and reducing density and thermal conductivity. This can be accomplished by carefully adjusting the amount of LWA and with a proper mix design to obtain reduced thermal conductivity and increased strength-to-density ratio (specific strength). It can be noticed from Table 5 that HGM incorporation in cementitious matrix leads to a reduced thermal conductivity coefficient comparable to the other LWA and an improved specific strength which is comparable to fly ash cenosphere (FAC) and significantly higher than the other LWAs, which makes it an ideal material for structural LWC applications. The information in Table 5 also shows that the compressive strength of LWC produced with natural aggregates such as pumice and perlite generally has lower compressive strength compared to those with artificial LWA (i.e. FAC and HGM) due to the fact that the latter have higher particle strength compared to the former. The use of expanded polystyrene (EPS) beads and polystyrene granules also results in significant reductions in compressive strength, which restricts the use of these materials where strength is also a major parameter.

Table 5 - A summary of LWC properties with various LWAs

Type of LWA	Reference	w/b	Dry Density (kg/m³)	Thermal conductivity (W/mK)	Compressive strength, 28d (MPa)	Specific strength (kNm/kg)
HGM	Present study	0.50	1670 - 2033	0.75 - 1.38	42.7 - 57.7	25.6 - 27.9
	[2][1]	0.43	1434 - 2027	0.66 - 2.18	35.23 - 52.69	21.3 - 29.0[4]
EPS beads	[2][1]	0.43	1300 - 2085	0.71 - 2.50	12.73 - 43.59	9.8 - 20.9[4]
Polystyrene granules	[51][1]	0.55	1560 - 1980	0.27 - 0.61	19.0 - 37.0[3]	12.2 - 18.7[4]
Fly ash cenosphere (FAC)	[52][1]	0.70	1098	0.41	23.54	21.4[4]
	[2][1]	0.43	1396 - 2019	0.80 - 2.21	35.4 - 53.5	23.1 - 26.5[4]
Expanded perlite (EP)	[53][2]	0.55	354 - 1833	0.13 - 0.60	0.1 - 28.8	0.3 - 15.7[4]
	[54][2]	0.36	945 - 1540	0.30 - 0.67	15.6 - 29.1	16.1 - 18.9
Pumice	[55][2]	0.64-1.23	1150 - 1271	0.345 - 0.455	14.63 - 26.09	12.7 - 20.5[4]
	[56][2]	0.48	1370 – 2370[3]	0.41 - 2.0[3]	10.0 - 51.0[3]	7.3 - 21.5[4]
Expanded glass	[57][2]	0.38 -0.59	1280 - 1490	0.485 - 0.847	23.3 - 30.2	18.2 - 20.3
Polyethylene beads (PEB) and Scoria+PEB	[58][2]	0.45	1366 - 1744	0.338 - 0.510	16.93 - 26.53	11.9 - 15.2[4]

[1] *Mortar mixes* / [2] *Concrete mixes* / [3] *Predicted values from the reference* / [4] *Calculated values*

Table 6 presents the test results of selected LWC mixes from Table 5 incorporating different types of LWA with similar densities (1660 ± 40 kg/m³) to compare their thermal insulation and strength properties.

Table 6 - Comparison of physical and mechanical properties of LWACs with similar density

Type of LWA	Reference	w/b	Dry Density (kg/m³)	Thermal conductivity (W/mK)	Compressive strength, 28d (MPa)	Specific strength (kNm/kg)
HGM	Present study	0.50	1670	0.75	42.7	25.6
HGM	[2]	0.43	1670	0.98	48.4	29.0*
EPS beads	[2]	0.43	1705	1.42	26.1	15.3*
Scoria+polyethylene beads	[59]	0.45	1621	0.392	20.9	12.9*
FAC	[2]	0.43	1669	1.15	62.8	37.6*
EP	[53]	0.55	1677	0.57	17.3	10.3*

* *Calculated values*

The present study shows that lightweight composite with an oven-dry density of 1670 kg/m^3, thermal conductivity coefficient of 0.75 W/mK, the compressive strength of 42.7 MPa, and specific strength of 25.6 kNm/kg can be manufactured by using HGM as LWA. With relatively low strength LWA, such as EPS beads [2] and expanded perlite [53], at similar density, the compressive strength and specific strength is 26 and 17 MPa and 15 and 10 kNm/kg, respectively. It can be seen from Table 6 that the higher specific strength could be obtained by using LWAs such as HGM and FAC, however, the lowest thermal conductivity values belong to mixes incorporating EPS beads, expanded perlite, and combination of scoria and polyethylene beads as LWAs. The results of the present study compared to the other studies in the literature confirm that strength reductions can be lowered by using HGM in LWC and also show its beneficial effects on the thermal conductivity, density, and specific strength.

3.4. Microstructure and Fracture Topography

The secondary scanning electron microscopic images of the fracture surface of paste mixes are shown in Figs. 13 and 14. As it is clearly seen in Fig. 13, HGMs are well distributed in the cement matrix as individuals without any agglomerations even at 40% incorporation ratio. In addition, it can be seen that the HGMs remained unreacted in the matrix. It can also be noticed that HGM-II mostly acts as weak points within the matrix which may promote the initiation and percolation of cracks (see Fig. 13 (c)). This phenomenon might have altered the physical and mechanical properties (Figs. 10-12) of mortar samples as the volume fraction of HGM increases, the mechanical properties decrease at higher dosages of HGM incorporation. The low mechanical strength of HGM-II particles could not resist the stresses within the matrix and the stress cracks mostly propagated through them, see Figs. 13 (d) and 14 (c). On the other hand, HGM-I particles mostly presented a different behaviour. The HGM-I particles are generally smaller in size (with an average particle size around 20 μm) and have higher crushing strength as compared to the HGM-II particles. Thus, the higher mechanical strength of HGM-I particles would alter the damage mode, where it can be observed that the crack growth was resisted by the HGM-I particles by hindering its path (Figs. 13 (a) and (b)). While some particles did break, many particles within the mortar have shown debonding instead of shell breakage, see Fig. 13 (a).
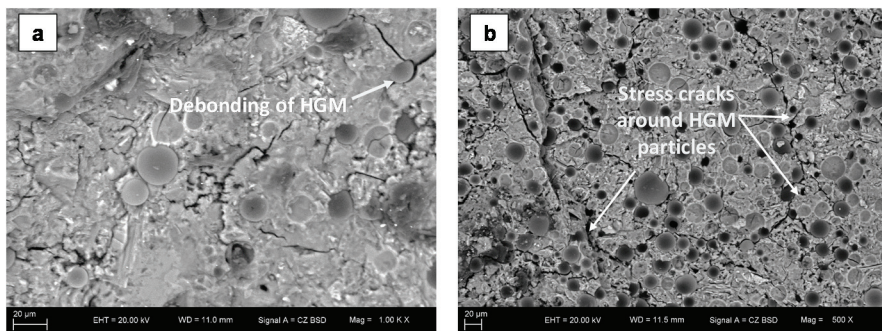
Fig. 13 - SEM micrographs of (a) HGM-I-10 (b) HGM-I-40 (c) HGM-II-10 (d) HGM-II-40 mixes
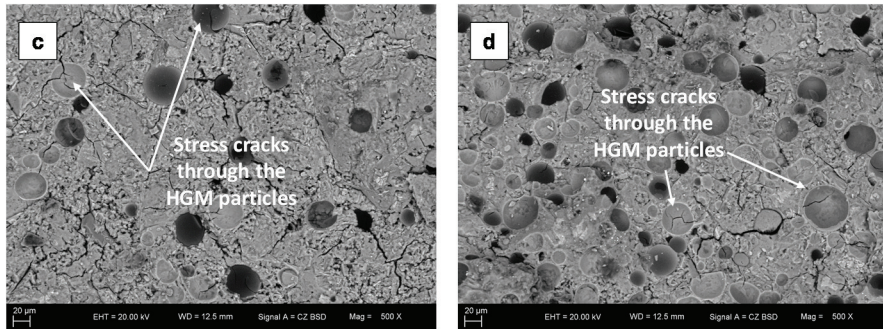
Fig. 13 - SEM micrographs of (a) HGM-I-10 (b) HGM-I-40 (c) HGM-II-10 (d) HGM-II-40 mixes (continue)

Fig.14 shows the interfacial transition zone characteristics of HGMs and the matrix. Generally, the dense cement hydration reaction products around the HGM particles are observed for both types of HGMs, which might also indicate that the distributed fine HGM in the cement matrix may act as nucleation sites for cement hydration products. However, large HGM-II particles allow stress cracks to propagate through their shell (Fig. 14 (c)) whereas the smaller HGM-I may act as barriers to hinder the propagation of stress crack (Fig. 14 (a)).
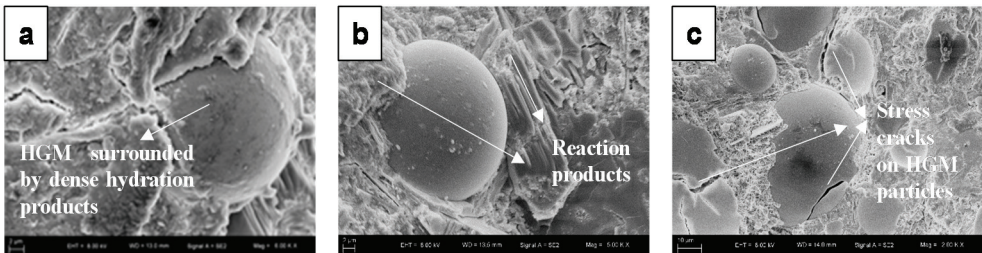

Fig. 14 - Microstructure of cement pastes incorporating HGMs: (a) HGM-I-10, (b) HGM-II-10, (c) HGM-II-40

3.5. Correlations between Physical and Mechanical Properties

It is well known that the physical and mechanical properties of concrete are correlated with each other and empirical relations have been developed between density, thermal conductivity, and compressive strength as shown in Tables 7 and 8. Since there is no available data in the literature which shows the relationship between density-thermal conductivity and density-compressive strength of HGM-modified cement composites, a regression analysis was performed to evaluate possible correlations between these parameters. Fig. 15 shows that the thermal conductivity and the compressive strength of HGM-modified composites can be predicted using the oven-dry density values with R-squared values of 0.9403 and 0.9116 respectively. The equation developed for thermal conductivity assessment was chosen in the

same format as the other works in the literature and presented comparable R^2 values as seen in Table 7. It should be noted here that for different LWA content and/or water to cement ratios, and for density values that fall outside the tested range of the present study, the equations proposed here should be used with care.

Table 7 - Empirical relations between density and thermal conductivity of lightweight concrete

Reference	Equation	R^2
Present study	$\lambda = 0.0718e^{0.0014\rho}$	0.9403
[56]	$\lambda = 0.0676e^{0.0015\rho}$	0.9498
[54]	$\lambda = 0.064e^{0.0015\rho}$	0.9476
[58]	$\lambda = 0.0625e^{0.0015\rho}$	0.8100
[60]	$\lambda = 0.000201\rho + 0.0776$	0.9440

Table 8 - Empirical relations between density and compressive strength of lightweight concrete

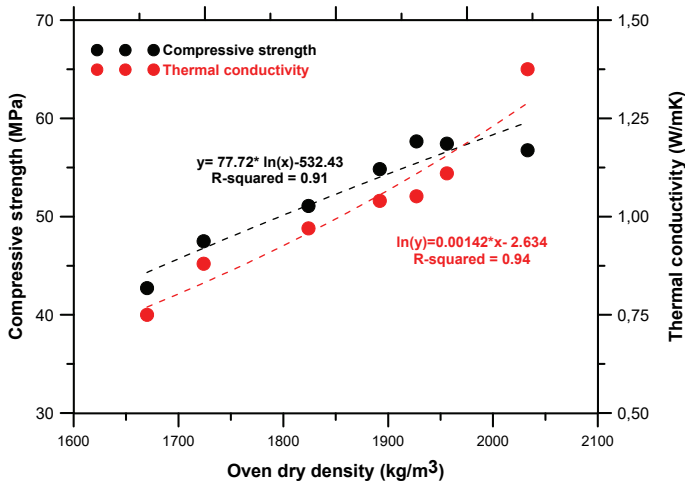| Reference	Equation	R^2
Present study	$f_c = 77.706\ln(\rho) - 532.31$	0.9116
[56]	$f_c = 4 \times 10^{-8}\rho^3 - 0.0002\rho^2 + 0.2734\rho - 128.64$	0.9100
[61]	$f_c = 0.1752\rho - 282.34$	0.8814
[62]	$f_c = 2 \times 10^{-17}\rho^{5.77}$	0.9710
[63]	$f_c = 10.3 \times \rho^{1.918} \times 10^{-6}$	0.9760

Fig. 15 - Correlations between compressive strength – oven dry density and thermal conductivity – oven dry density

4. CONCLUSIONS

Low thermal conductivity and density are the main characteristics of lightweight cement-based composites. Producing lightweight composites with improved thermal, physical, rheological, and mechanical properties is still a great concern for energy-efficient buildings. In this study, lightweight cementitious mortars were developed with improved thermal and physical characteristics by using HGMs. It was found that the composites comply with the ACI specifications of structural lightweight concrete which requires density between 1120 - 1920 kg/m^3 and a minimum compressive strength of 17 MPa at 28 days. To observe the effectiveness of the different types of HGMs on the rheological, physical, thermal, and mechanical characteristics, flow diameter, yield stress, thermal conductivity, along with the compressive and flexural strength tests were performed.

Based on the findings of this study, the following outcomes may be drawn:

- The particle size of the HGMs was found to be one of the most important parameters affecting the rheological properties and the flow diameter of the cement matrix. The finer the HGM resulted in higher yield stress and plastic viscosity and lower flow diameter. The HGM incorporation ratio also affected the rheological properties and flow diameter of cement paste and mortar, a consistent reduction in workability was noted with an increasing amount of both types of HGMs.

- The incorporation of HGM is found to be an effective method to obtain lower density and thermal conductivity. The type and amount of HGM play an important role in the reduction of thermal conductivity and density, HGM-II mixes possessed lower density and thermal conductivity values. Moreover, higher contents of HGM provided higher reductions. It was found that the density and the thermal conductivity of cement mortar may be reduced by up to 20% and 45%, respectively, by 40% HGM inclusion.

- The incorporation of HGM altered the mechanical properties of cement mortars. The compressive strength of cement mortar slightly increased with both HGMs at 10% ratio. The 20% HGM ratio was found to be critical for compressive strength where only minor reductions were observed compared to the reference, beyond this ratio (at 40%) the compressive strength reduced by 16% and 25% in HGM-I-40 and HGM-II-40 mixes, respectively. However, the compressive strength of mortars with 40% HGM inclusion may still satisfy the requirements of most civil engineering components in practice, since the compressive strength values appeared to be more than 43 MPa.

- The HGM-I mixes showed higher strength values than the HGM-II mixes, due to the higher mechanical strength characteristics compared to that of HGM-I. This also indicates that HGMs with higher mechanical strength should be chosen to achieve cement composites with higher mechanical properties.

- Comparison with other works revealed that at similar composite density, HGM as a LWA contributes to mechanical properties of cement composites more than the natural LWAs and EPS and polyethylene beads. Strong correlations were established between oven-dry density, thermal conductivity, and compressive

strength of HGM-modified composites, with correlation coefficients comparable to other studies.

- The SEM micrographs indicate homogeneous, well-distributed HGMs in the matrix and presence of dense cement hydration reaction products around the HGM particles. It was also noticed that HGM-I incorporated mixes presented a better performance in terms of resisting to stress cracks, which was the key parameter affecting the mechanical behavior.

It can be seen from the overall test results that the HGM-incorporation has a great influence on the thermal and physical performance of cement mortar mixes. The incorporation of HGM provides lower thermal conductivity and density. Moreover, desirable compressive strength values can be obtained by adjusting the amount of HGMs. It is thought that the results of the present study may contribute to the literature and also in the design and development of lightweight cementitious composites with enhanced thermal and physical properties by the incorporation of HGMs.

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