

Mechanical and thermal properties of lightweight geopolymer composites

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Abstract

This research has investigated the properties of thermally insulating geopolymer composites that were prepared using waste expanded polystyrene as lightweight aggregate. The geopolymer matrix was synthesized using metakaolin and an alkaline activating solution. To improve its mechanical properties, this matrix was modified by the addition of an epoxy resin to form an organic-inorganic composite. Moreover, in order to reduce drying shrinkage marble powder was used as an inert filler. The materials obtained were characterized in terms of physico-mechanical properties, thermal performance and microstructure. The geopolymer expanded polystyrene composite have improved properties compared to Portland cement-based materials, with higher strengths and lower thermal conductivity. The research demonstrates the manufacture of sustainable lightweight thermally insulating geopolymer composites using waste expanded polystyrene.

Keywords: Expanded polystyrene, geopolymer, composite, thermal insulation.

1. Introduction

Expanded polystyrene (EPS) is an extremely lightweight thermoplastic that has low thermal conductivity, high durability and low-cost. EPS is widely used in many thermal insulation applications and as lightweight packaging [1]. The end of life recycling and reuse options for EPS are limited and it is normally either landfilled or incinerated. This can cause environmental problems in countries where appropriate standards are not enforced [2]. Several recycling processes have been developed for EPS [3], but these often require the use of hazardous solvents [4]. This research has investigated using waste EPS as a lightweight aggregate in metakaolin derived geopolymer. The objective was to develop lightweight thermally insulating materials with mechanical properties suitable for use in non-structural applications. At the same time, a recycling option for EPS that allows this material to remain in the economic cycle is provided through use in new sustainable materials. Waste EPS has reduced environmental impact compared to many other types of waste derived manufactured lightweight aggregates [5-11].

Previous research has investigated EPS in Portland cement composites [12-25]. These studies report that a substantial decrease in compressive strength is associated with increasing the EPS content, and this requires the addition of materials, such as silica fume and steel fibres to improve mechanical performance. The properties of EPS concrete depend on the mix design and the EPS particle size distribution [26]. Increased shrinkage and creep deformation are reported and result from a reduction in the restraint effect compared to natural aggregates, which have much higher static modulus of elasticity [27-30]. Additional issues related to EPS lightweight aggregate concrete are Eigen stress-driven cracking and increased bulk shrinkage [31]. EPS-containing concrete has reduced spalling resistance at high temperature due to thermal decomposition of EPS [18]. The embedded CO₂ is increased with EPS addition due to the high carbon content of EPS compared to normal inorganic cement binders and aggregates.

Several strategies have been proposed for reducing the embedded CO₂ in the built environment [32-33]. Geopolymers are innovative binders that have been extensively researched in recent years consisting of amorphous aluminosilicates that are synthesized using alkaline activation of solid precursors such as fly ash [34-36], calcined clays [37-40] and blast furnace slag [41-43]. Geopolymers are a potential alternative to traditional Portland cement in selected applications, because they combine reduced environmental impact with excellent mechanical properties. However, they have relatively low toughness

and low flexural strength and in order to improve these properties geopolymer composite materials have been formed by the *in situ* co-reticulation of a geopolymer matrix with an epoxy based organic resin [44–49]. These modified geopolymer materials show enhanced compressive and flexural strength compared to normal geopolymers with analogous compositions due to the synergistic effects between the inorganic and the organic phases arising from interfacial forces at nanometre scale. The properties are controlled by composition and processing method and these modified geopolymer materials have potential to be used in structural [50], photo-catalytic [51], fire-resistant and thermal insulating [52, 53] applications.

Lightweight geopolymers have been prepared with different mix proportions by foaming [54] and using different lightweight aggregates [55-61]. In this research, lightweight geopolymer concrete (LWGC) has been investigated using recycled EPS as aggregate. Geopolymer matrix preparation used metakaolin (MK) and an alkaline activating solution (AAS). Epoxy resins with tailored composition and stoichiometry were added to obtain geopolymer organic composites. Waste calcium carbonate powder from processing marble has been used as a filler as this improves the mechanical properties of geopolymers and reduces drying shrinkage [63]. This waste is a major problem that effects the environment [63]. The LWGC samples prepared were tested for physico-mechanical and thermal properties and the interfacial zones between EPS particles and the geopolymer matrix characterised by microstructural analysis.

2. Materials and methods

2.1 Materials

The composition of metakaolin (MK, Neuchem S.r.l.) sodium silicate solution (SS, Prochin Italia S.r.l) and marble powder [64, 65] are shown in Table 1. Reagent grade sodium hydroxide was supplied by Sigma-Aldrich and the epoxy resin (Epojet®) was supplied by Mapei S.p.A. EPS was obtained from a waste treatment plant in Campania, Italy and consisted of <5 mm particles with an apparent density of $1.6 \pm 0.3 \times 10^{-2} \text{ g/cm}^3$. The EPS was from polystyrene seed trays used in agriculture and these were processed by milling to produce EPS beads. Waste marble slurry was dried at 105 °C for 4 hours and milled to produce marble powder (MP) with particle sizes ranging between 10 and 300 µm.

Table 1

Chemical composition (weight %) of the metakaolin (MK), marble powder (MP) and sodium silicate solution (SS).

	Metakaolin	Marble powder	Sodium silicate
SiO ₂	52.90	1.12	27.40
Al ₂ O ₃	41.90	0.37	-
CaO	0.17	52.26	-
Fe ₂ O ₃	1.60	0.11	-
MgO	0.19	0.87	-
K ₂ O	0.77	0.10	-
Na ₂ O	-	0.14	8.15
Water	-	-	64.45
LoI	-	40.74	-

*LoI= Loss on Ignition

The compositions of the LWGC mixes are given in Table 2. The alkaline activating solution was prepared by dissolving solid sodium hydroxide into the sodium silicate solution. The solution was then allowed to equilibrate and cool for 24 hours. The composition of the solution can be expressed as Na₂O·1.4SiO₂·10.5H₂O. Geopolymer pastes were obtained by mixing MK for 10 minutes with the activating solution, at a solid to liquid ratio of 1:1.4 by weight, using a Hobart mixer. EPS beads and MP were then added and the system mixed for a further 5 minutes. This procedure was used for the LWGC samples that did not contain epoxy resin. These were the GMK-65, GMK-MP-65, GMK-72.5 and GMK-MP-72.5 mixes. GMK- XX samples contained EPS, where XX refers to the amount of EPS v/v%. GMK- MP-YY samples are sample containing EPS and MP, where YY refers to the sum of EPS and MP v/v%.

Epoxy resin geopolymer composites (GMK-E10-XX and GMK-E10-MP-YY) were produced by adding 10 w/w % by weight of Epojet® resin to the freshly-prepared geopolymer suspension and mixing for 5 minutes. Epojet® resin was cured at room temperature for 10 minutes before adding to the geopolymer mix when it was workable and before cross-linking and hardening had occurred.

After mixing the pastes were cast into prismatic (40 x 40 x 160mm) and cubic (100 x 100 x 100 mm) moulds and cured sealed at 40°C for 24 hours. The specimens were kept

sealed at room temperature for 6 days and then stored in air at room temperature for a further 21 days.

Table 2

Composition (weight %) of the materials prepared in this research.

Sample	MK	SS	NaOH	Resin	MP filler*	EPS beads*	
						Wt.	Vol.
GMK-65	41.6	50.0	8.4	-	-	1.9	65.0
GMK-MP-65	41.6	50.0	8.4	-	7.5	1.7	63.3
GMK-72.5	41.6	50.0	8.4	-	-	2.8	72.5
GMK-MP-72.5	41.6	50.0	8.4	-	7.5	2.8	70.8
GMK-E10-65	37.4	45.0	7.6	10	-	1.9	65.0
GMK-E10-MP-65	37.4	45.0	7.6	10	7.5	1.7	63.3
GMK-E10-72.5	37.4	45.0	7.6	10	-	2.8	72.5
GMK-E10-MP-72.5	37.4	45.0	7.6	10	7.5	2.8	70.8

*Calculated with respect to geopolymer paste and/or geopolymer composite (with resin) paste.

2.2 Methods

The apparent density of samples was determined as the ratio of the mass to a given volume by hydrostatic weighing using an OHAUS-PA213 balance. The compressive and flexural strengths were evaluated according to EN 196-1. The tests were performed after 28 days curing and the values reported are the average of six strength tests. Flexural strength tests on prismatic samples used a Controls MCC8 multipurpose testing machine with a capacity of 100 kN. Compressive strength measurements on cubic samples used a Controls MCC8 hydraulic console with 2000 kN capacity. Thermal conductivity tests were performed on 100 x 100 x 100 mm cube samples using a Hot Disk M1 analyser (Thermal Instruments Ltd). This is a non-destructive test based on the transient plane source technique according to ISO 22007-2:2015. Microstructural analysis by scanning electron microscopy (SEM) used a Phenom Pro X Microscope on freshly prepared fracture surfaces. Optical images were obtained from polished surfaces.

3. Experimental results and discussion

3.1 Morphological characterization

Figure 1 is a SEM image of an EPS particle showing the typical cellular structure [66].

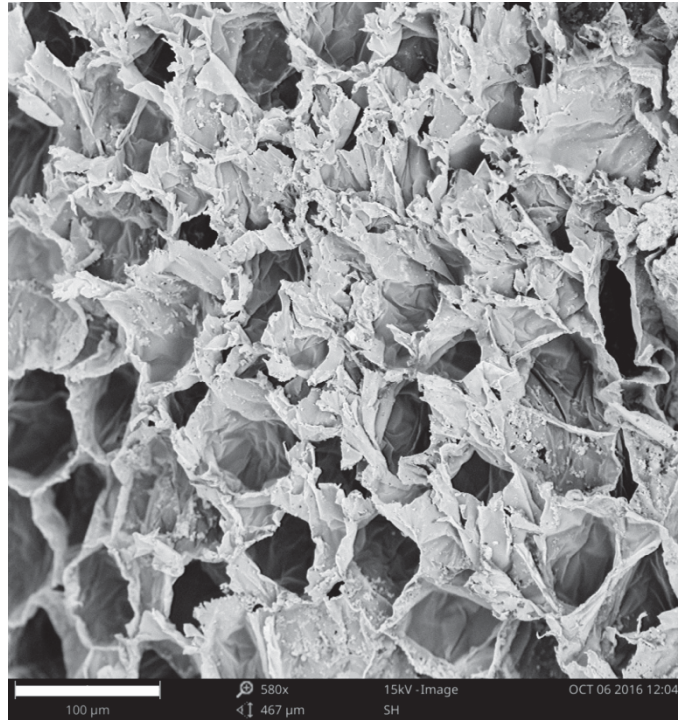


Fig. 1. SEM image of an EPS particle. Scale bar is 100μm

Due to the grinding process, these cells are not evenly distributed and vary in dimensions.

Figure 2 shows optical micrographs of polished surfaces of GMK-72.5 and GMK-E10-72.5 samples.

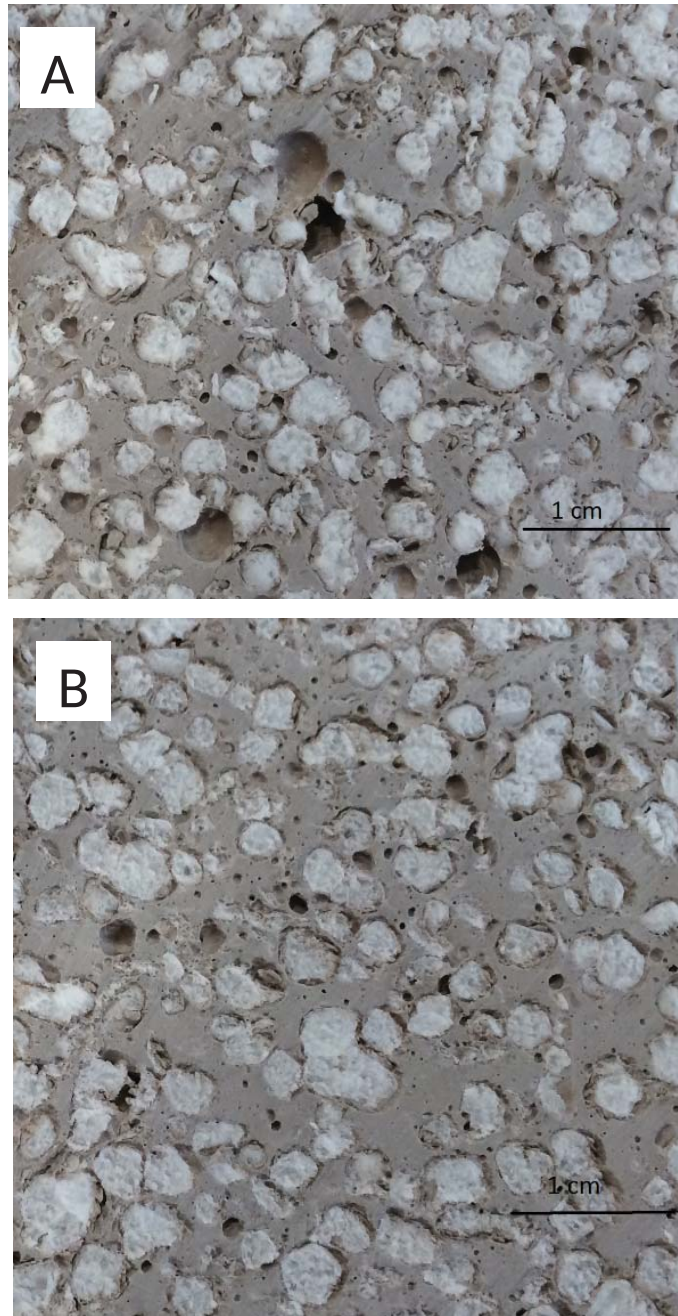


Fig. 2. Optical micrograph of polished surfaces of A) GMK-72.5 and B) GMK-E10-72.5.

The EPS beads are embedded in the geopolymer matrix and distributed uniformly with no evident aggregation phenomena. Moreover, the specimens show a compact structure with no cracking, as confirmed by SEM images of these samples that was used in order to investigate in detail the microstructure of the samples and the bonding characteristics between the geopolymer matrix and EPS particles and MP aggregate (Figure 3). This demonstrates that at microscopic level, the matrix is compact and homogeneous. The SEM images in Figures 3 (A and A', sample GMK-72.5) indicate that there is very good

1 adhesion between EPS particles and the matrix. EPS particles are completely embedded in
2 the geopolymer and it is difficult to clearly identify the interface. This compatibility was
3 obtained without the use of any additives.

4 The adhesion between EPS particles and the matrix is also good for samples
5 prepared using the composite matrix containing epoxy resin (Figure 3B, B', sample GMK-
6 E10-72.5). The major difference is in the matrix microstructure, which shows the presence
7 of microspheres of resin of various sizes as discussed in our previous work [47].

8 The addition of MP (Figure 3C, C', sample GMK-E10-MP-72.5) as filler does not
9 compromise the bonding between phases in the geopolymer matrix thus not affecting
10 significantly the microstructure. The particles are well dispersed and the strong adhesion
11 improves the mechanical properties.

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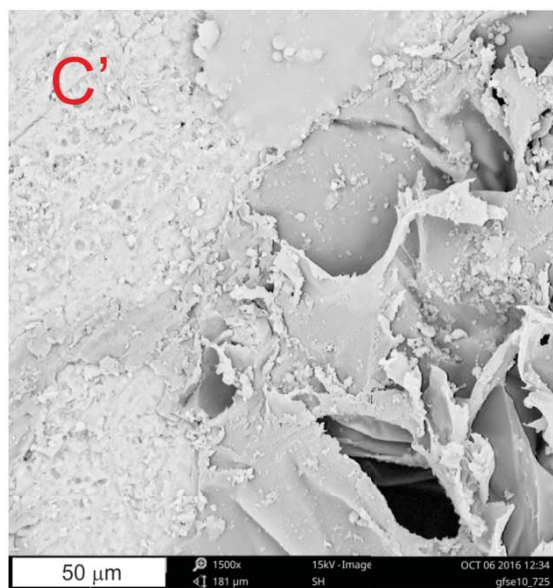
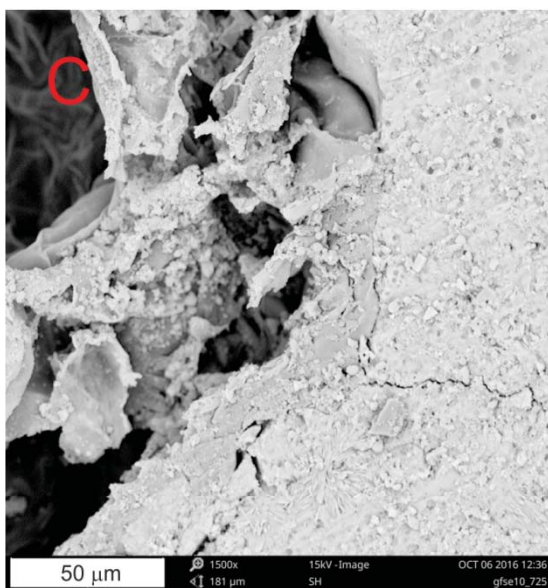
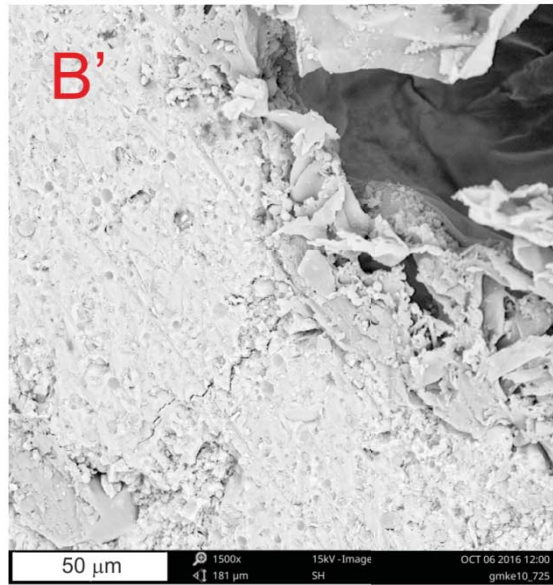
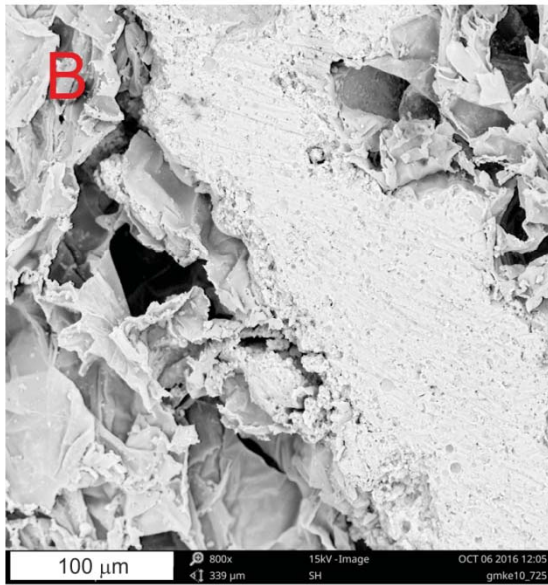
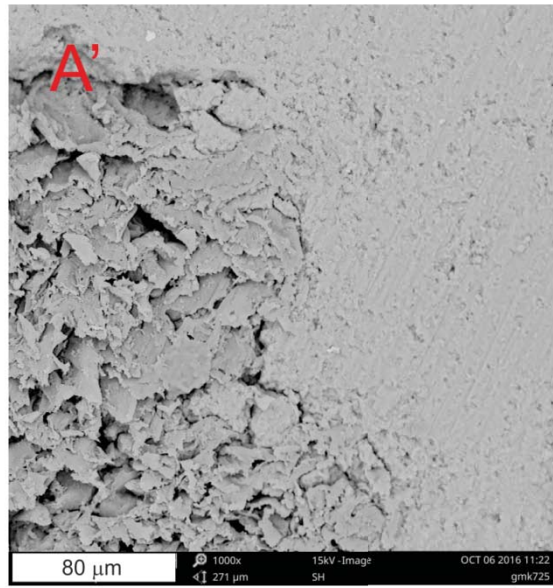
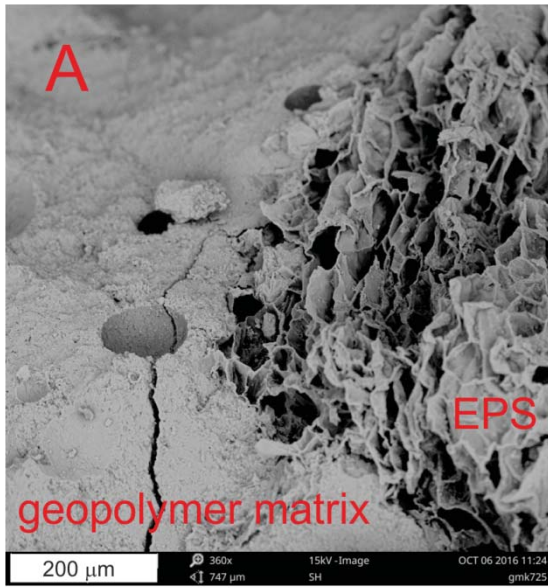


Fig. 3. SEM images of an interface area between an EPS particle embedded in the geopolymer matrix: **A, A')** neat geopolymer matrix (sample GMK-72.5); **B, B')** composite geopolymer matrix (sample GMK-E10-72.5); **C, C')** composite geopolymer matrix containing also marble powder (sample GMK-E10-MP-72.5). In all cases a very good adhesion between EPS particles and the matrix is apparent.

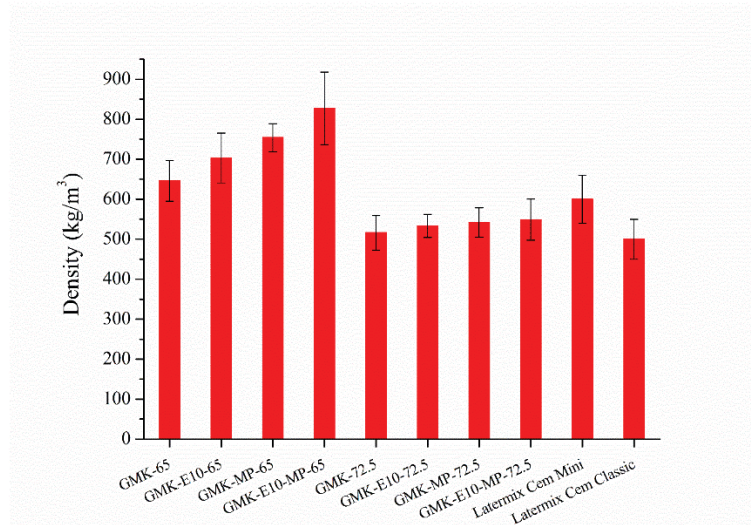
3.2 Physico-mechanical characterization.

Figure 4a shows the apparent density of samples. As expected, density decreases as the content of EPS aggregate increases. Samples with 65% volume of aggregates had densities ranging from $646 \pm 51 \text{ kg/m}^3$ (GMK-65) to $827 \pm 91 \text{ kg/m}^3$ (GMK-E10-MP-65). Samples with a 72.5 % volume content of aggregates had densities ranging from $516 \pm 43 \text{ kg/m}^3$ (GMK-72.5) to $549 \pm 52 \text{ kg/m}^3$ (GMK-E10-MP-72.5). For neat geopolymer samples (GMK-65 and GMK-72.5), increasing the volumetric content of EPS by less than 10% turns out in a decreased of the density by $\sim 20\%$. More pronounced decreases in density were observed for the samples containing epoxy resin and MP. In particular, correspondingly to the same increase of EPS content, the samples with epoxy resin in the geopolymer matrix (GMK-E10-65 and GMK-E10-72.5) showed a decrease of density $\sim 24\%$, while in the case of the addition of MP (GMK-MP-65 and GMK-MP-72.5), the decrease of density is $\sim 27\%$. Finally, in the case of the addition of both organic resin and MP (GMK-E10-MP-65 and GMK-E10-MP-72.5) the decrease of density is $\sim 33\%$. Moreover, from the data reported in Figure 1, it is apparent that the organic resin and MP additions have a more limited influence on the density of samples containing 72.5% EPS in respect to those at lower EPS content (for example, the addition of the organic resin and MP turns out in an increase of density of $\sim 28\%$ in the case of the samples with 65% vol of EPS beads and of only 6% in the samples with 72.5% vol of EPS).

The geopolymer samples had comparable densities to EPS-containing Portland cement matrices [14] and commercial EPS-containing concrete mixtures for which values around 1000 kg/m^3 are reported [67]. The mechanical performance of EPS-containing geopolymer concrete correlates with density. The volumetric content of aggregate influences both compressive and flexural strengths (Figure 4b, c). The compressive strengths (Figure 4b) of LWGC samples containing 65% volume of EPS beads ranged from 3.4 ± 0.5 to $6.0 \pm 1 \text{ MPa}$, while for higher EPS volumes (72.5%) compressive strengths ranged from 1.8 ± 0.3 to $2.4 \pm 0.2 \text{ MPa}$. It is apparent that the addition of both marble powder and epoxy resin significantly improved the mechanical properties of

samples. The best compressive strength values were obtained for specimens GMK-E10-MP-65 and GMK-E10-MP-72.5, and the values obtained were comparable to commercial alternatives [67] and greater than the literature data on EPS-containing Portland cement composites.

A similar trend to compressive strength was observed for flexural strength (Figure 4c). For EPS contents of 65% the flexural strength varied from 0.32 ± 0.08 MPa for geopolymer samples to 0.6 ± 0.1 MPa for composite matrix samples with MP. With greater EPS contents (72.5%) the flexural strength ranged from 0.22 ± 0.07 to 0.33 ± 0.09 MPa and only a minor improvement in mechanical properties was associated with the addition of MP and epoxy resin. It could be argued that in these samples with higher EPS content, the very poor mechanical properties and high compressibility behaviour of polystyrene particles neutralize the beneficial effect on the mechanical properties of the addition of epoxy resin and MP (that instead is evident in the set of samples with lower EPS content) by causing the formation of micro-cracks at the interface between the geopolymer matrix and the EPS particles.



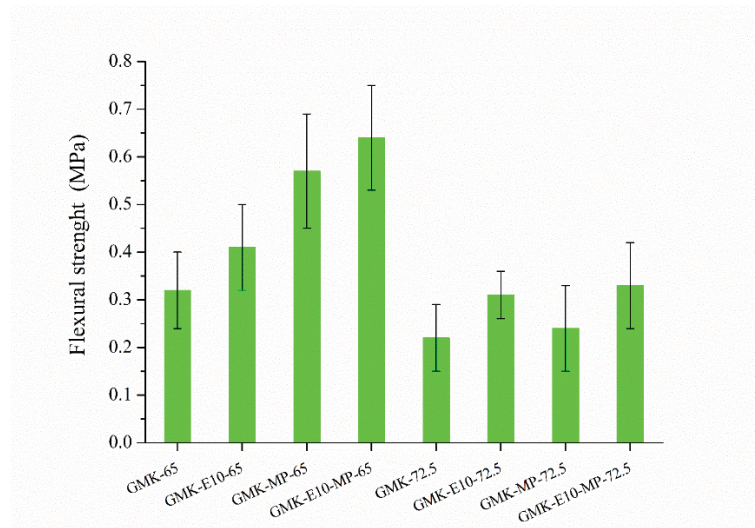
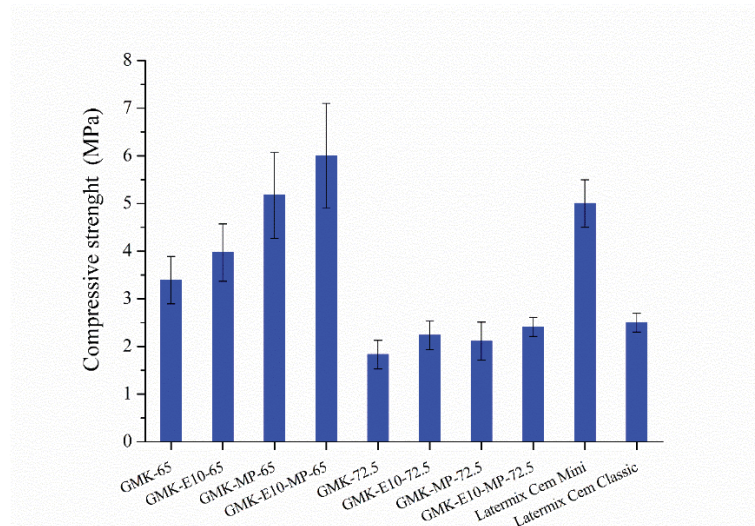


Fig.4: Apparent density (a), compressive strength (b) and flexural strength (c) of LWGC samples prepared. In a) and b), the data for two commercial products (Latermix Cem Mini© and Latermix Cem Classic©, <http://www.laterlite.es/wp-content/uploads/2014/03/General-Catalogue.pdf>) are also reported for comparison.

3.3 Thermal properties

Figure 5 shows thermal conductivity data for the LWGC samples prepared in this study. As for density data (Figure 4a), two different groups of specimens can be identified. Samples containing 65 v/v% of aggregates had greater thermal conductivity than the samples containing 72.5 v/v% of EPS. For example, sample GMK-65 had a thermal conductivity of 0.158 ± 0.001 W/m·K while sample GMK-72.5 had a thermal conductivity of 0.121 ± 0.001 W/m·K, a 23.4% reduction. It is apparent that, as expected, the presence of EPS particles causes a significant reduction in thermal conductivity. The correlation between thermal conductivity and density for LWGC samples is shown in Figure 6. The

samples with the highest thermal conductivity (0.207 ± 0.001 W/m·K) was sample GMK-E10-MP-65 which had the highest bulk density (827 ± 91 kg/m³), while the sample with the lowest thermal conductivity (0.121 ± 0.001 W/m·K), sample GMK-72.5, had the lowest density. The influence of MP and epoxy resin on thermal conductivity is not clear as these are minor components in the samples tested.

The addition of MP and epoxy resin to geopolymers produced LWGC with significantly improved mechanical properties compared to lightweight mortars made with Portland cement with similar thermal conductivity. For example, sample GMK-72.5 retained good mechanical properties and had very low thermal conductivity (0.121 ± 0.001 W/m·K). This is 15% lower than Portland cement based commercial products with similar density. [67]. The reduction in thermal conductivity increases to 92% when compared to analogous materials with the same density that had poor mechanical properties compared to the samples prepared in this study [19].

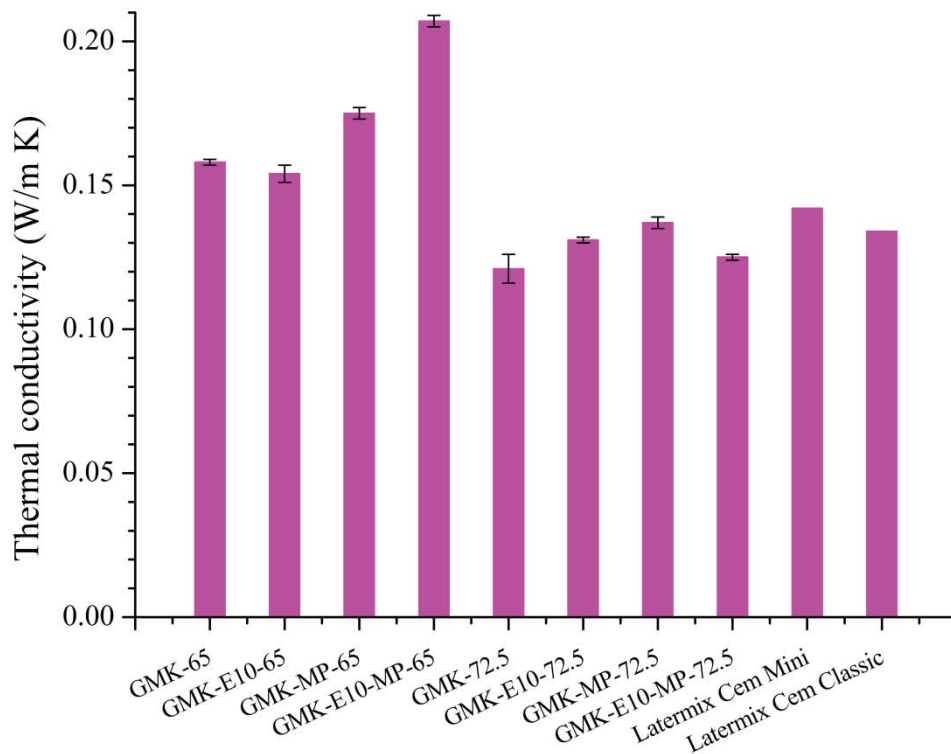


Fig. 5. Thermal conductivity of LWGC samples. Data for two commercial products (Laternix Cem Mini© and Laternix Cem Classic©), are also reported for comparison. [67]

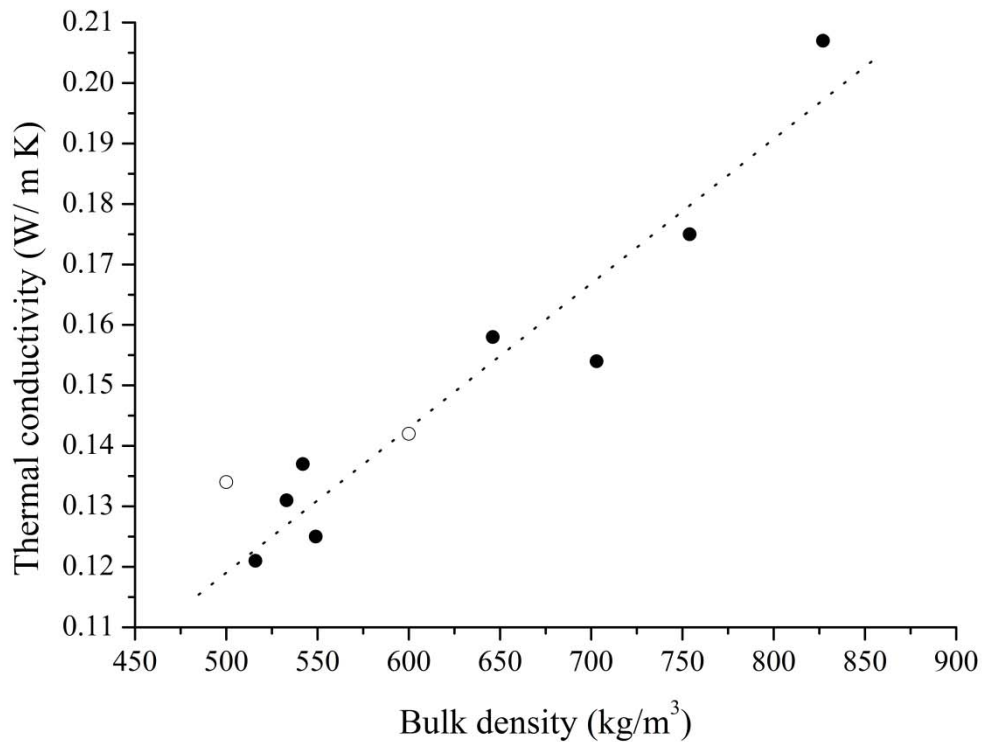


Fig. 6. Correlation between thermal conductivity and density of LWGC samples: full circles (●) are related to LWGC samples; empty circles (○) are related to two commercial products (Latermix Cem Mini© and Latermix Cem Classic©, [67]).

4. Conclusions

Lightweight thermally insulating materials based on geopolymer concrete containing expanded polystyrene (EPS) as insulating aggregate were prepared and characterized. The microstructural characterization showed a homogeneous structure with EPS beads uniformly dispersed and embedded in the geopolymer matrix. Compressive and flexural strengths decreased with increasing EPS content. The addition of an organic resin to the geopolymer significantly increased both compressive and flexural strengths. A similar effect was observed with the addition of marble powder. All samples studied were characterized by very low thermal conductivity. This was much lower than analogous lightweight materials with similar densities reported in the literature. The research has demonstrated the production of geopolymer matrix EPS composites that are lightweight thermally insulating materials with excellent mechanical properties.

Acknowledgements

The authors thank Neuwendis S.p.A. for the metakaolin supply and Prochin Italia S.r.l. for the silicate solution supply. Mr. Giovanni Morieri e Mrs. Luciana Cimino are warmly acknowledged for assistance in laboratory activities. Università di Napoli “Parthenope” is acknowledged for financial support with a grant within the call “Support for Individual Research for the 2015-17 Period- Annuity 2016” issued by Rector Decree no. 954 (28/11/2016).

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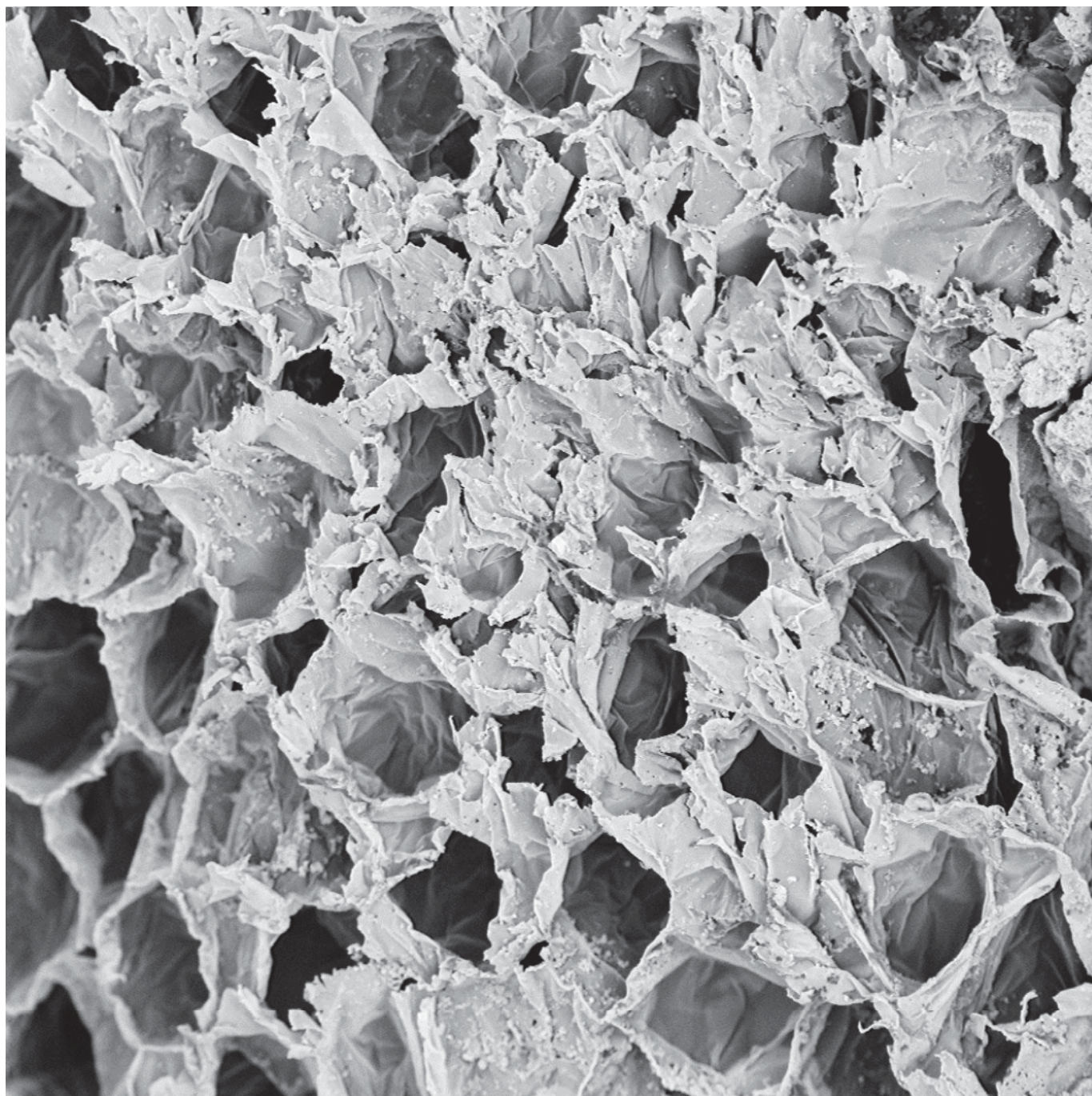
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