PRODUCTION OF LIGHTWEIGHT FILLERS FROM WASTE GLASS AND PAPER SLUDGE ASH

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ABSTRACT:
The production of high-performance lightweight fillers (LWFs) has been investigated using recycled glass and paper sludge ash (PSA), as this represents a high value re-use application for these materials. PSA exhibits low sintering reactivity at temperatures below 1200 °C. However, milled glass undergoes liquid-phase sintering at temperatures where gases formed from the decomposition of calcium carbonate present in PSA are encapsulated. This produces lightweight porous materials suitable for use as LWFs. The effect of key process parameters such as PSA content, particle size of the raw materials and sintering conditions such as temperature and time, have been optimised. Processing glass containing 20 wt. % PSA at 800 °C produces particles with appropriate physical and mechanical properties that are comparable to leading commercially available LWF products.

Keywords: Lightweight fillers; Recycled glass; Wastepaper sludge ash; Liquid phase sintering; Construction materials

1. Introduction

The construction industry is the largest consumer of natural, non-renewable resources, and has a significant impact on the sustainability of the UK and other countries worldwide. The supply of raw materials to be used in the manufacturing of high-performance construction materials is increasingly under pressure. Depleting natural sources of raw materials and increasing energy costs are key drivers towards increased sustainability in the construction sector. In order to address both sustainability and competitiveness challenges that have arisen from stricter legislation and the adverse economic climate, an increasing trend is the use of wastes as resources to substitute for natural materials [1]. The production of artificial secondary aggregate and filler materials would provide an alternative with both environmental and economic benefits. This reduces reliance on quarrying primary materials and diverts wastes from landfill. Engineering secondary aggregates with high recycled content will also help to meet the increasing demand for construction aggregates [2].

The development of new civil infrastructure is important in many countries and is a key driver for economic growth. This is driving innovation in the construction sector as projects increasingly need to deliver complex architectural designs with high-performance and efficiency targets, which require the use of versatile materials. High-rise buildings and structures void or unstable ground, tunnelling in urban areas and earthquake-vulnerable

regions are critical cases where reducing the dead-load of structures has advantages. The use of lightweight building materials is necessary to meet these challenges.

The production of lightweight fillers (LWFs) from recycled glass and paper sludge ash (PSA) has been identified as a promising reuse application given that LWF materials have higher value than normal weight or lightweight aggregates (LWAs) currently in use. In comparison with lightweight aggregates, LWFs have lower density and water absorption rates and are supplied in sizes typically ranging from 0.5 mm to 4 mm diameter. LWFs provide low-thermal conductivity, sound proofing properties and potentially improved fire-resistance in additional to being light-weight.

Artificial LWFs are formed by rapid sintering at high temperatures using able to generate gas that produces appropriate levels of residual porosity. Two requirements have to be met during sintering of materials [3]:

- the evolution of gases coming from the thermally unstable constituents;
- presence of a highly viscous liquid phase to allow the encapsulation of gases.

Recycled glass is suitable for the formation of a viscous phase. It has high silica content, has an amorphous structure and large surface area when milled, and has been successfully used for the production of lightweight materials [4-6]. Due to the high sodium oxide (Na_2O), calcium oxide (CaO) and silicon dioxide (SiO_2) content, glass has quite low sintering temperature, and this reduces the firing time and energy consumption [7, 8]. Sodium in the glass is responsible for the formation of a low-viscosity melt that is able to encapsulate the evolving gases.

Expanded glass particles can therefore be produced by mixing finely ground glass with a suitable expanding agent and firing this mixture at a temperature above the softening point of glass where the viscosity is less than $10^{6.6}$ $N \cdot s \cdot m^{-2}$ [9]. Amongst various expanding agents, PSA has been selected as a source of CO_2 gas coming from the decomposition of calcium carbonate. PSA is currently used in the manufacture of blocks, as an animal bedding material and it is applied to agriculture land as a liming agent [10]. Therefore the manufacture of glass-PSA LWFs constitutes a novel, higher-value application for the increasing amounts of PSA that are likely to be produced. This represents an alternative to PSA disposal via landfill which occurs in the UK and elsewhere [11].

The objective of this work was to prepare glass-PSA LWFs and characterise their physical and mechanical properties. These were compared with commercially available LWFs imported from Germany that are increasingly being used in the UK.

2. Experimental

2.1 Materials

2.1.1 Glass cullet

Mixed-colour recycled glass cullet was used. This was ground using attrition milling for 60 seconds in 50g batches to produce a fine glass powder which was used in all the experiments. Particle size of glass powder was <100 μm. The absolute density, measured using a helium gas pycnometer (Micromeritics, AccuPyc II 1340, Georgia, USA), was 2.48 $g.cm^{-3}$. The chemical composition of recycled glass was determined by X-ray fluorescence (XRF Spectro 2000 Analyser) as shown in Table 1.

2.1.2 Paper sludge ash

Paper sludge ash (PSA) was supplied by a major paper mill in South East England, producing newsprint from recycled paper. They use combustion in a fluidized bed boiler at 850 °C to manage waste paper sludge. The PSA produced is a fine grey powder with pH~12. The chemical composition is presented in Table 1.

The particle size distribution of as-received PSA samples was determined by laser diffraction (Beckman Coulter LS-100). PSA has a d_{10} (10% by volume of particles having a diameter smaller than this size) of 23 μm, d_{50} (mean diameter) of 156 μm and d_{90} of 395 μm. The absolute density, excluding the pores between particles, measured using a helium gas pycnometer (Micromeritics, AccuPyc II 1340, Georgia, USA), was 2.85 $g.cm^{-3}$.

The crystalline phases present in PSA were determined by X-ray diffraction (XRD, Philips PW 1830 diffractometer system with 40 mA and 40 kV, Cu radiation). As shown in Figure 1, the major crystalline phases present are gehlenite ($Ca_2Al_2SiO_7$), mayenite ($Ca_{12}Al_{14}O_{33}$), calcite ($CaCO_3$), calcium silicate (a'–Ca_2SiO_4), lime (CaO) and quartz (SiO_2). PSA particles are porous and heterogeneous as shown by the SEM micrographs in Figure 2. Separate particles form larger agglomerates as a consequence of combustion.

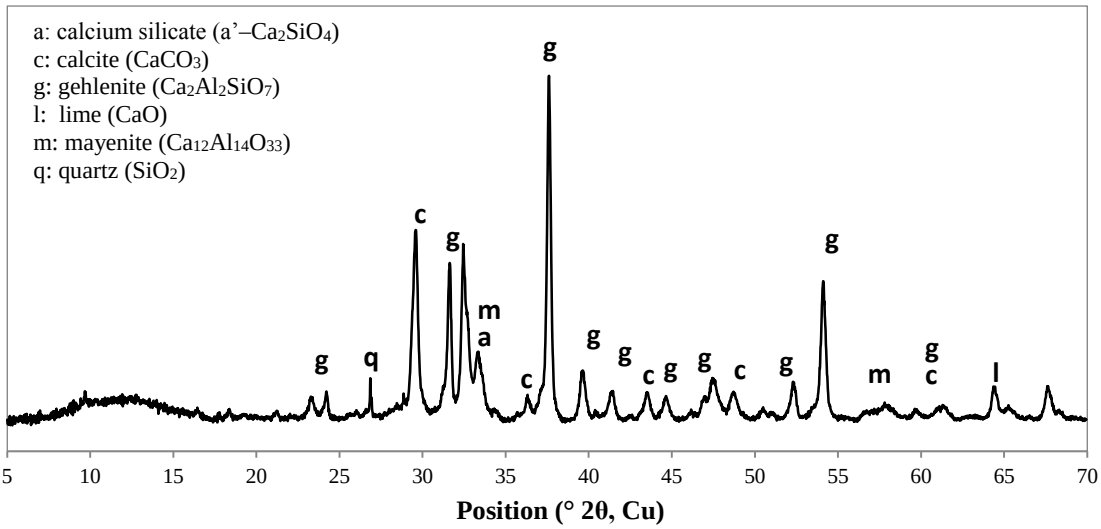

Figure 1. X-ray diffractogram of as-received paper sludge ash.

Table 1. Chemical composition (wt. %) of recycled glass and PSA

	SiO_2	CaO	Na_2O	MgO	Al_2O_3	K_2O	Fe_2O_3	SO_3	TiO_2	P_2O_5	Others
Waste glass	75.8	12.0	7.3	2.3	1.4	0.6	0.3	0.2	nd	nd	-
PSA	21.2	61.2	nd	2.8	12.6	0.4	0.9	0.2	0.3	0.1	0.1

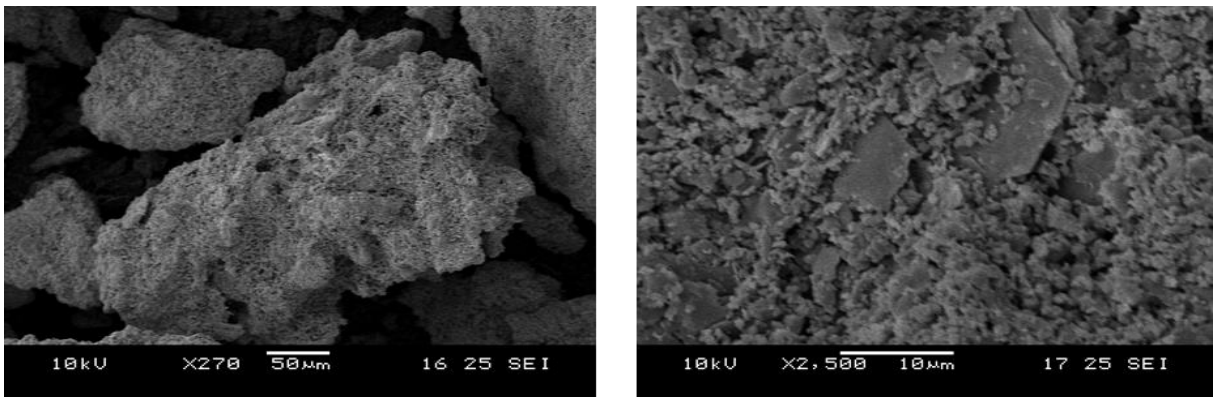

Figure 2. Scanning electron micrographs of PSA particles

2.1.3 Characterisation of sintering behavior

The sintering behavior of recycled glass and PSA has been characterized using dilatometry (Netzsch 402E). Pressed fine glass powder and PSA samples were heated to 800 °C and 1400 °C respectively at a constant rate of 10 °C/min. Thermogravimetric analysis of

PSA (Netzsch STA 449 F1 Jupiter®) used dried powder weighing ~35mg with a heating rate of 10 °C/min.

2.2 Preparation of PSA-glass lightweight fillers (LWFs)

Milled recycled glass was mixed with various amounts (0-50 wt.%) of PSA by wet ball milling (Pascal Engineering Ltd.) at a constant rate of 45 rpm for 1 hour using 19 mm diameter alumina balls as the milling media. 500g batches of raw materials were used with 1000 mL of water with a solid: milling media ratio of 1: 5. Wet-milling resulted in thick slurry which was dried at 105 °C for 24 hours. The dried glass-PSA powder was manually ground using a pestle and mortar and sieved to < 475 μm. Spherical particles were formed using a pan-pelletiser with the addition of ~50% (w/v) water. Green pellets ranging in diameter from 0.5–5 mm were fired in a Carbolite rotary tube furnace. This had a 150 cm long tube with a 90 cm central heated zone. The speed of rotation was 10 rpm and it was kept horizontal to control the sintering time.

2.3 Characterisation of sintered products

The dry density of the sintered LWFs (apparent specific gravity) was determined using Archimedes Principle [12]:

$$\rho_{\mathrm{rd}} = \rho_{\mathrm{w}} \cdot \frac{\mathrm{m_{dry}}}{\mathrm{m_{sat} - m_{imm}}}\,\mathrm{g\,cm^{-3}},$$

where the dry mass is m_{dry}, immersed mass is m_{imm} and 24-h saturated surface-dry mass is m_{sat}. In order to ensure full saturation, samples were kept under water and under vacuum for 24 hours.

The water absorption (WA_{24}) was calculated according to the following formula:

$$\mathrm{WA_{24}} = \frac{\mathrm{m_{sat} - m_{dry}}}{\mathrm{m_{dry}}} \times 100\%.$$

The compressive strength of LWFs produced was determined by loading a bulk amount of 3-4 g of LWFs between two parallel plates until 10% deformation is achieved. The confined compressive strength CS (10) was calculated using [13]:

$$\mathrm{CS(10)} = \frac{\mathrm{F_{10}}}{\mathrm{A}},$$

where F_{10} is the load recorded at 10% deformation, and A is the area of the load distribution plate.

The physical properties (particle density (n=10), water absorption (n=10) and confined compressive strength (n=10)) of commercially produced LWFs have also been determined using the same test methods.

Fracture surfaces of pellets were Au coated and examined using scanning electron microscopy (SEM, JEOL JSM-5610LV) to investigate the microstructure.

3. Results and discussion

3.1 Sintering properties of recycled glass and PSA

The dilatometer data for PSA and glass are presented in Figure 3. Recycled glass powder exhibits shrinkage of 23.2% between 594 °C and 664 °C, which indicates the temperature range within which glass softening occurs. This is comparable data to that

reported by Karamanov et al. [14]. PSA has low reactivity as particles did not sinter together to form a rigid body and samples fell apart after heating to 1180 °C.

3.2 Thermogravimetric analysis of PSA

Thermogravimetric analysis data for PSA is shown in Figure 4. TGA of PSA shows an initial weight loss between ambient and 120 °C associated with the evaporation of residual water. Weight loss at higher temperatures is caused by thermal degradation of PSA. Weight loss between 600 °C and 780 °C is attributed to the decomposition of calcite ($CaCO_3$) to lime (CaO) with simultaneous generation of CO_2. Evolution of CO_2 gas within this temperature range makes PSA a potential expanding agent for preparing porous glass particles.

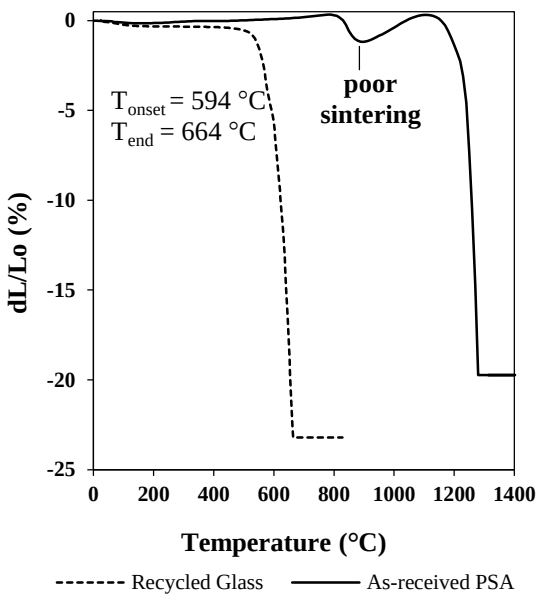

Figure 3. Dilatometer results for glass and PSA.

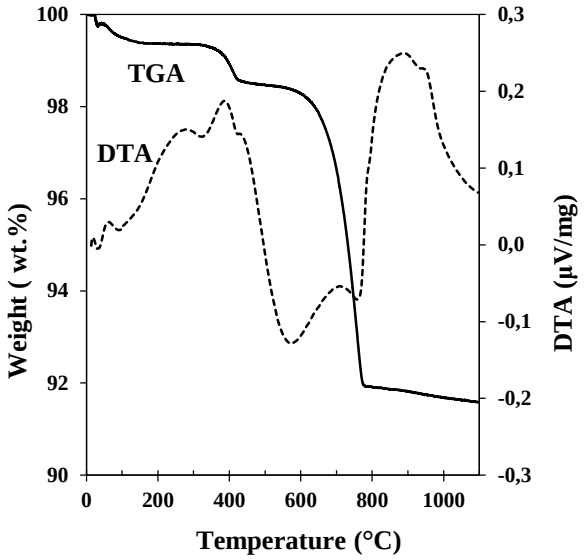

Figure 4. TGA and DTA results for PSA.

3.2 Physical and mechanical properties of sintered products

3.2.1 Effect of PSA addition on glass mixes

The effect of PSA addition on density and water absorption of various glass-PSA mixes is shown in Figure 5 which also contains data for a commercial LWF.

Samples prepared with PSA additions up to 20 wt. % have similar or lower densities than 5 mm commercial LWFs. Further increases in the PSA content result in increased density. At 10 wt. % addition of PSA, lightweight fillers with a particle density as low as 0.45 g cm^{-3} are obtained. However, excessive interconnection of pores and piercing of the vitrified layer formed due to the presence of glass results in high water absorption of 145 %. The optimum samples, combining low density and relatively low water absorption, contained 80 wt. % glass and 20 wt. % PSA and these had a density of 0.75 g cm^{-3} and water absorption of 75 %. In contrast, glass sintered pellets have a particle density of 1.12 g cm^{-3} and water absorption of 3.8 %.

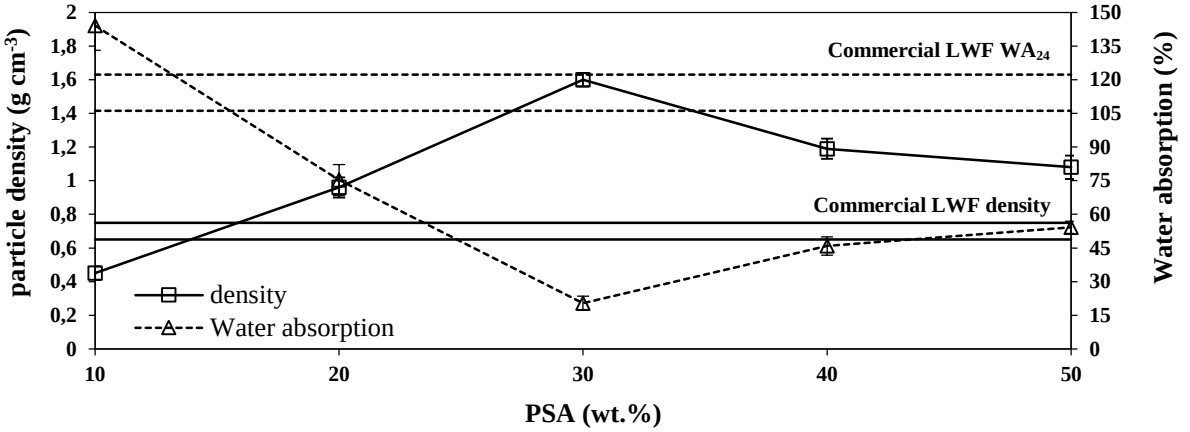

Figure 5. Effect of PSA addition on the density and water absorption of sintered glass-PSA pellets at 800 °C.

3.2.2 Effect of sintering time on physical properties of LWFs

The effect of sintering time on the 80/20 glass/PSA pellets fired at 800 °C is shown in Figure 6. Microstructural evolution can be described by four distinct stages: a) heating, b) glass softening/gas evolution, c) stabilisation and d) further densification. The expansion process of sintered LWFs is a dynamic balance between the evolution of gaseous species from PSA and the inhibiting formation of viscous glass layers able to encapsulate those gases.

Based on this data, the optimum sintering time was 15 minutes with LWFs having a density of 1 g·cm^{-3} and WA of 17%. The limitations on pore size increase can be explained by the disintegration of the rigid cell walls of sintered particles reflected on the rapid increase in water absorption rates when sintering between 15 and 20 minutes. After sintering for 20 minutes, no significant changes were observed in the LWF properties tested.

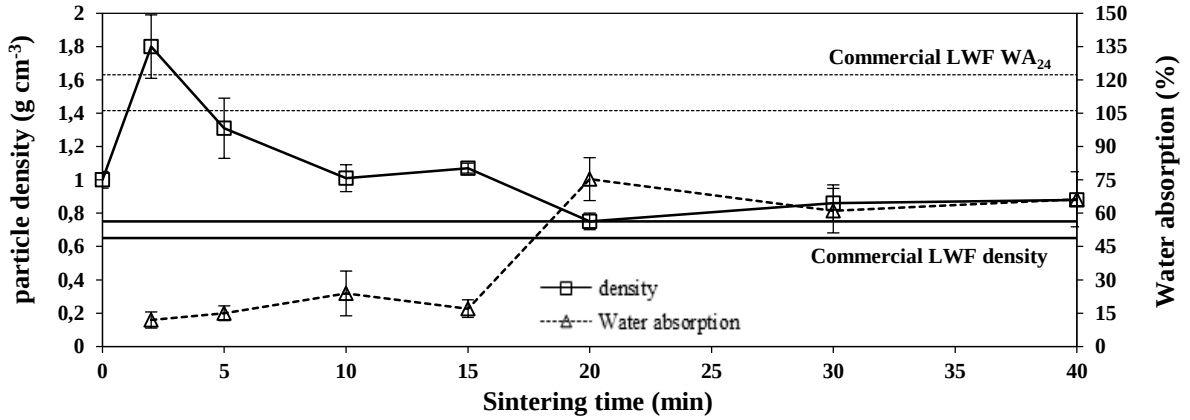

Figure 6. Effect of sintering time on density and water absorption of 80/20 glass/PSA mixes sintered at 800 °C.

3.2.3 Microstructure of LWFs

The fracture surfaces of the optimum 80/20 glass/PSA LWFs and commercial LWFs are shown in Figure 7. This shows that a uniform distribution of approximately spherical pores with typical diameters of 50-150 μm is formed in glass/PSA pellets fired at 800 °C for 15 minutes, as opposed to a network of 10-40 μm pores present in pure glass sintered pellets. Interconnection of pores was engineered to a minimum by controlling the sintering time. Formation of rigid walls between the pores ensures improved mechanical behaviour of the LWFs produced.

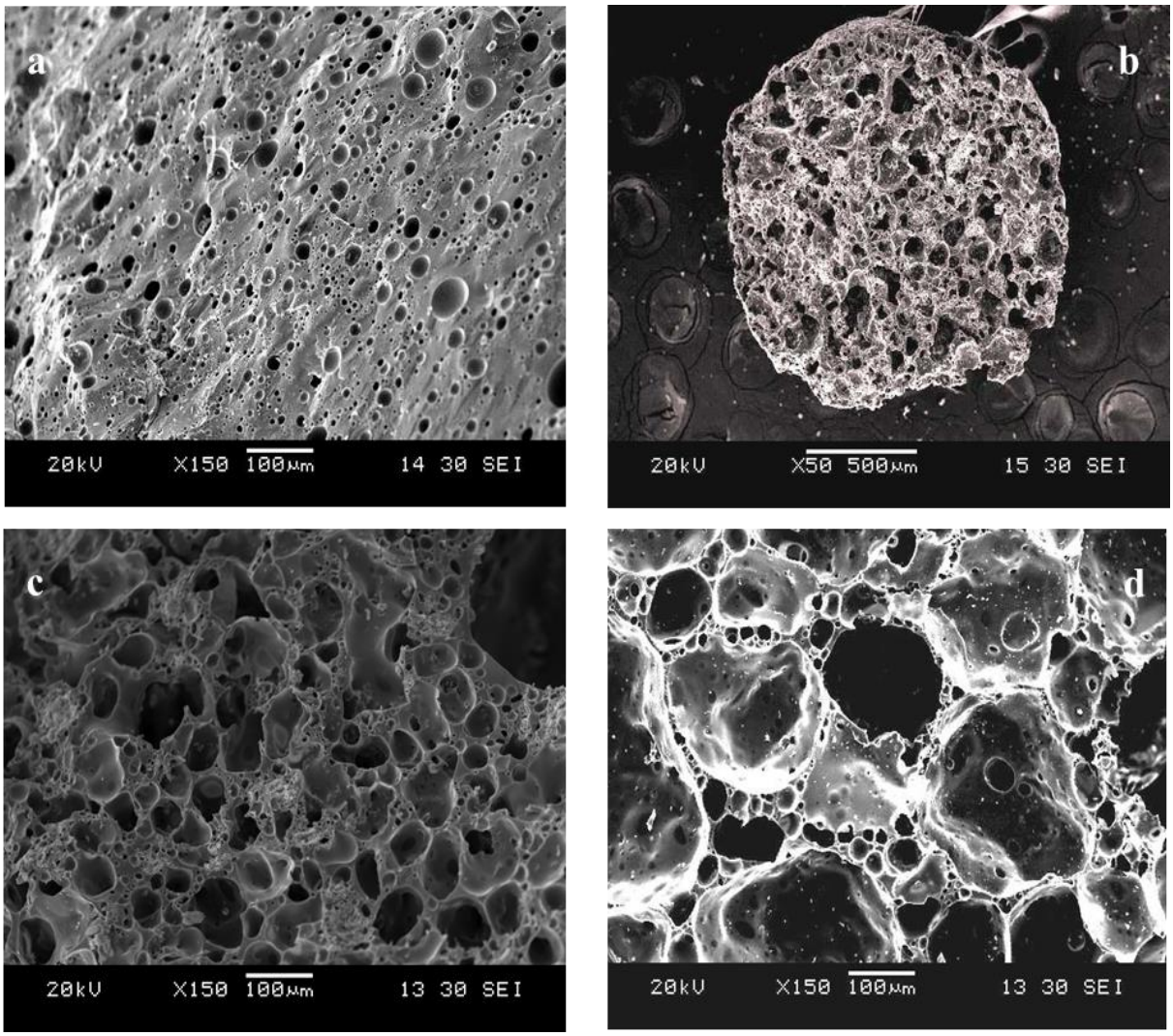

Figure 7. Morphology of LWFs tested: a) Cross-section of glass sintered pellets at 800 °C for 15 minutes, b) Sintered 80/20 glass/PSA LWF sintered at 800 °C for 15 min. c) Higher magnification image of sample depicted in (b), d) Commercial LWF.

3.2.4 Mechanical properties of LWFs

The confined compressive strength CS(10) for 80/20 glass/PSA LWFs sintered at 800 °C for 15 minutes with a diameter of 1-2 mm and 2-5 mm are compared against commercial LWFs in Table 2. Artificial glass-based LWFs with 20% addition of PSA have consistent mechanical properties regardless of the LWF size. The crushing strength is 2.9 MPa and this is three times higher than values for typical commercially available LWFs.

Table 2. Mechanical properties of 80/20 glass/PSA LWFs and commercial LWFs

Sample ID	CS (10) (MPa)
Commercial LWF 1-2 mm	1.3
80/20 glass/PSA LWFs 1-2 mm	2.9
Commercial LWF 2-5 mm	1.1
80/20 glass/PSA LWFs 2-5 mm	2.9

4. Conclusions

The object of this work was to develop lightweight filler particles using waste glass and paper sludge ash (PSA). The conclusions derived from the results are:

- A glass-PSA system can form lightweight materials using simple processing technology involving wet milling, pelletising and low temperature sintering;
- addition of recycled glass aids sintering of PSA given the low sintering reactivity of PSA;
- encapsulation of evolving gases within the particle body during sintering is the predominant mechanism responsible for the LWF microstructure;
- lightweight fillers containing 80 wt. % glass and 20 wt. % PSA sintered at 800 °C for 15 minutes have density of ~1 g cm^{-3} and water absorption of 17 % compared to 115 % for the commercial LWFs tested;
- the results indicate potential for the production of high-performance LWFs with mechanical properties significantly enhanced compared to commercial products.

Acknowledgements

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