Properties and microstructure of lightweight aggregate produced from lignite coal fly ash and recycled glass

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Abstract

The effect of glass addition on the processing, physical properties and microstructure of lightweight aggregate made from lignite coal fly ash from the Megalopolis power station in Greece has been investigated. Fly ash/glass mixes have been rapidly sintered at temperatures between 1040 and 1120 ºC in a rotary furnace, and the density, water absorption and pellet strength determined. Sintering 60: 40 fly ash: waste glass mixes at 1120 ºC produced lightweight aggregate with a mean density of 1.35g/cm^3, water absorption of ~16% and crushing strength of 7.3 MPa. Major crystalline phases in sintered materials were quartz (SiO_2), albite ($NaAlSi_3O_8$), moissanite (SiC), hematite (Fe_2O_3), wollastonite ($CaSiO_3$) and diopside ($CaMg(Si_2O_6)$). The work indicates that Megalopolis fly ash combined with waste glass can be used to manufacture lightweight aggregate with properties comparable to commercially available products. Fly ash and glass are potential resources that are currently waste materials in Greece. The processing involving pelletising and sintering in a rotary kiln is similar to that required for other commercially available lightweight aggregates manufactured from shales, clays and slate, and therefore processing costs are expected to be similar. However, avoiding the costs and environmental impacts associated with importing lightweight aggregate or using pumice makes the production of FA/glass lightweight aggregate a viable option.

Keywords: Lightweight aggregate; Fly ash; Sintering; Glass; Coal combustion products; Lignite

1. Introduction

Coal fired power stations in Greece produce approximately 10,000 kilo tonnes of fly ash (FA) per annum (Skodras et al. 2007). The highest level of production is in West Macedonia in northern Greece, where approximately 7,600 kilo tonnes is produced each year. In Peloponissos in the southern part of Greece, about 2,400 kilo tonnes/year is produced. Only a relatively small amount of FA in Greece is currently beneficially reused. The cement industry uses between 7 and 15% of total FA production, but FA reuse in Greece is generally low compared to much of Europe, where typically 48% is reused (Quina et al. 2003).

During the last two decades, significant scientific research has been completed in Greece, to increase the use of FA. This work has characterized FA (Filippidis and Georgakopoulos 1992; Georgakopoulos et al. 1994; Sakorafa et al. 1996; Fytianos et al. 1998; Georgakopoulos et al. 2002; Arditsoglou et al. 2004; Karangelos et al. 2004; Koukouzas et al. 2006; Skodras et al. 2007) and assessed potential applications including use in cement and concrete (Papayianni and Valiasis 1991; Kouloumbi and Batis 1992; Papayianni 1993; Kouloumbi et al. 1994; Malami et al. 1994; Harris and Moehlum 2003; Papayianni and Anastasiou 2003; Antiohos and Tsimas 2005; Papadakis and Tsimas 2005; Papagianni 2005; Papayianni and Milud 2005; Voglis et al. 2005; Skodras et al. 2007). The use of FA in zeolite production (Mouhtaris et al. 2003; Moutsatsou et al. 2006) and road construction (TEE 1988; AUTH 1994; Mouratidis 2001; Kehagia and Tsoxos 2005) have been reported. The potential for using FA in cement production, brick manufacture and construction of road embankments in Greece has also recently been investigated (Skodras et al. 2007). It was concluded that total FA utilisation in these applications would be less than 40% of the total assuming optimistic reuse levels. The development of new applications for lignite coal FA is therefore necessary in order to reduce reliance on landfill. National specifications for the utilisation of FA came into effect in 2007 (Greece 2007). This is an additional driver for the development of new applications that provide solutions to the FA management problem in Greece.

The European Union Directive 94/62/EC sets the legislative framework related to packaging waste (EUROPA 2007). The Directive covers all packaging waste including glass, paper and paperboard, plastics and wood and sets the requirements that all EU Member States have to meet regarding the reusable and recoverable nature of packaging and packaging waste. It also requires Member States to take measures, which may include national programmes, to prevent the formation of packaging waste, and encourages them to develop packaging reuse systems.

A major component in municipal solid waste (MSW) is container glass, even though it is relatively easy to separate (Karamberi and Moutsatsou 2005). Glass is included in the EU Packaging Directive and specific recycling rates have been set for Member States. By 2011, 60% by weight of glass will have to be recycled in Greece. Waste glass generation and utilization rates in Greece for the period between 1997 and 2005 are presented in Table 1. This shows that the waste glass recycling rates between 2000 and 2005 were at about 26% (EUROPA 2007).

There is a limit to the amount of glass cullet that can be re-used in glass production due to contamination by impurities, and therefore there is a surplus of green cullet in Greece (Papadopoulos 2003a; Papadopoulos 2003b; Enviros 2004). The lack of markets means that alternative uses are required to reduce the amount sent to landfill. Glass has been successfully used as a binder and fluxing agent in ceramics and bricks, as it lowers the softening temperature, firing time and energy consumption (Barbieri et al. 1997; Nagaraj and Ishikawa 1999; Barbieri et al. 2000; Ducman et al. 2002; Karamberi and Moutsatsou 2005).

There is increasing demand for lightweight construction products for use in pre-cast, structural panels and multi-storey buildings. The most frequently used lightweight aggregates are made from expanded clay, shale, perlite, vermiculite and different kinds of sintered waste (Ducman et al. 2002). A range of different waste materials have been investigated for producing lightweight aggregate including incinerator bottom ash (IBA) (Bethanis and Cheeseman 2004; Quina et al. 2006), fly ash (Terukina et al. 1993; Swan et al. 2001; Huang et al. 2007) and glass (Ducman et al. 2002). Pumice is quarried primarily from the island of Gialli and this is currently the major type of lightweight aggregate used in Greece, with an annual production of ~800,000 tonnes. However, it is expected that future European legislation, driven by issues concerned with archaeological heritage in the area and the destruction of the natural environment on the island will mean that quarrying natural pumice will reduce and will eventually be stopped (Anagnostopoulos and Stivanakis 2009). The development of alternative artificial lightweight aggregates is therefore necessary to meet future domestic markets in Greece and provide a more sustainable source of lightweight aggregate.

The objective of this research was therefore to develop lightweight aggregate from lignite coal fly ash and waste glass and to characterise the properties and microstructure of the materials produced. The effects of sintering temperature on key physical and micro-structural characteristics relevant to use as lightweight aggregate are reported and compared to commercial lightweight aggregates.

2. Experimental

2.1 Materials

2.1.1 Fly ash

Type C fly ash (FA) was obtained from the Megalopolis power station (Public Power Corporation ΔEH). This burns lignite coal with low calorific value, high ash and high moisture content. The vast majority of the resulting FA is disposed of in old mines near Megalopolis. A representative batch of FA was collected and the chemical composition is compared with other FA samples from Greece in Table 2 (Nagaraj and Ishikawa 1999; TEE 2000; Quina et al. 2006; British_Glass 2007).

Major crystalline phases present in < 150 μm sieved Megalopolis FA were determined by x-ray diffraction (XRD, Philips P1830 diffractometer system using Cu K_{α} radiation). As shown in Figure 1, the main crystalline phases identified were quartz (SiO_2), gehlenite ($Ca_2Al_2SiO_7$), anorthite ($Ca_2Al_2SiO_8$), anhydrite ($CaSO_4$), hematite (Fe_2O_3) and calcite ($CaCO_3$). These minerals have previously been reported as major constituents in Megalopolis FA (Sakorafa et al. 1996).

Figure 2 shows that < 150μm particles of Megalopolis FA examined using scanning electron microscopy (SEM, JEOL JSM-5610LV) consist of irregularly shaped, roughly spherical particles of various sizes. Spherical particles are in the form of plerospheres (hollow particles filled with smaller particles or microspheres), cenospheres (hollow particles) and spherical particles < 5μm. Individual particles are often agglomerates of smaller particles and irregular shaped particles appear to be incompletely combusted carbonaceous material.

2.1.2 Glass cullet

Glass cullet was milled for 20 seconds in 100g batches to produce a fine powder in a Gy-Ro Mill® (Glen Creston Limited). Batches were thoroughly mixed to form a homogenous sample that was used in all experiments. Chemical composition data for glass cullet is included in Table 2.

2.2 Preparation of FA-glass lightweight aggregate (LWA) pellets

Less than 150 μm FA was thoroughly mixed with different amounts (0-40 wt %) of glass powder. An organic binder (1 wt% of Alcotac CB6, Ciba Speciality Chemicals) and 30% w/v water were added to produce a mix consistency that could be readily formed into roughly spherical 8 to 10mm diameter pellets. These were dried overnight at room temperature and sintered by rapidly passing them through an electric rotary tube furnace (Carbolite). This had an internal diameter of 7.7 cm, was 150 cm long, and had a 90 cm central heated zone. The speed of rotation was 2.8 revolutions per minute and the angle of tilt was held constant at 2°. A series of baffles were fitted inside the tube to control the pellet traverse along the length of the tube.

2.3 Characterization of sintered products

The dry density of the sintered aggregate pellets (apparent specific gravity) was determined using Archimedes Principle (Neville 1994):

$$\rho_{pellet} = \frac{m_{dry}}{m_{sat} - m_{imm}} \ g.cm^{-3}$$

where the dry mass is m_{dry}, immersed mass is m_{imm} and 24-hour saturated surface-dry mass is m_{sat}. The water absorption was calculated from:

$$Water\ Absorption = \frac{m_{sat} - m_{dry}}{m_{dry}} \times 100\%$$

The compressive strength of individual pellets was determined by loading individual pellets between two parallel plates and loading to fracture at P_c. The tensile strength is then given by (Hiramatsu 1965):

$$S = \frac{2.8 P_c}{\pi X^2}$$

where S is the tensile strength of the specimen, X is the distance between loading points. Five pellets were tested for each test condition to minimise statistical error.

Physical properties (particle density, water absorption and compressive strength) of commercially manufactured lightweight aggregates (Lytag, Aardelite and Optiroc) were also determined using identical test conditions to those for sintered FA-glass pellets. Crushed samples were ground to < 150 μm and the crystalline phases determined by XRD and fracture surfaces of samples were examined by scanning electron microscopy (JEOL JSM-5610LV).

3. Results

3.1 Sintered pellets physical properties

The effect of firing temperature on density and water absorption of 100% FA samples is shown in Figure 3. The samples were not effectively sintered as indicated by the high water absorption of ~53% which indicates excessive open porosity. All 100% FA samples were friable with inadequate physical properties for use as lightweight aggregate. Comparative data for commercial lightweight aggregates are included in Figure 3.

The effect of glass on fired density, water absorption and pellet strength is shown in Figure 4, for samples sintered at 1100 and 1120 ºC. Comparative data for commercial lightweight aggregates is also included. Glass provides soda (Na_2O), lime (CaO) and silica (SiO_2) to the mix composition and lowers the sintering temperature, resulting in increased density, reduced water absorption and increased compressive strength.

Figure 5 shows the effect of sintering temperature on fired density, water absorption and strength for FA with 20 and 40% glass additions. The densities are fairly constant at approximately 1.07 g/cm^3 for both sets of samples. This is lower than the density of Lytag but higher than other commercial lightweight aggregates such as Aardelite and Optiroc.

Samples containing 80% FA and 20% waste glass have high water absorption compared to the commercial products that were tested, with water absorption values between 34.4 and 38.2%. Samples containing 60% FA and 40% glass have lower water absorption values. The lowest value of 16.4% was obtained for the sample containing 60% FA and 40% waste glass fired at 1120 ºC. This was lower than the value obtained for Lytag and Aardelite.

The strength data shows that most samples had lower strengths than Lytag except the material made with 60% FA and 40% waste glass, fired at 1120ºC (7.4 MPa compared to 6.3 MPa for Lytag). All samples exhibited higher compressive strength than Aardelite and Optiroc and had increased strength at higher sintering temperatures.

3.2 Microstructure of sintered fly ash/glass

3.2.1 Mineralogy

XRD data obtained from the outer part of the 60% FA, 40% waste glass sample fired at 1120ºC is shown in Figure 6. This material consists of a complex composition with albite ($NaAlSi_3O_8$), hematite (Fe_2O_3), moissanite-3H (SiC), dolomite, ferroan ($Ca(Mg,Fe)(CO_3)_2$) and calcium (Ca) as the main crystalline phases. There is also some quartz (SiO_2), rutile (TiO_2) and graphite © present. A marked reduction in the quartz peak and the complete disappearance of calcite and plagioglace minerals that are present in the as-received FA occurs on sintering at 1120 ºC.

Figure 7 shows the XRD data for the inner part of the 60% FA and 40% waste glass sample fired at 1120 ºC. This sample displayed a black coring effect and the material analysed was from the central black core. The major crystalline phases identified were diopside ($CaMg(SiO_3)_2$), wollastonite-1A ($CaSiO_3$), augite ($Ca(Fe,Mg)Si_2O_6$), cristoballite (SiO_2) and albite, calcian ($(Na, Ca)Al(Si, Al)_3O_8$). The data shows the presence of significant amorphous glass as indicated by the high background level of the XRD data (Chimenos et al. 1999; Eusden et al. 1999).

3.2.2 Micro-structural examination

SEM images of fracture surfaces of samples sintered at 1100 ºC are presented in Figures 8. At 1100ºC the 100FA sample is not properly sintered (Figure 8a). The micrograph of the aggregate containing 20% waste glass indicates a denser more sintered microstructure. The addition of glass lowers the sintering temperature and promotes sintering, although large open pores still remain. The fracture surface of the sintered material containing 40% waste glass shows a dense matrix containing some isolated, approximately spherical pores that are typically 10 to 50 μm in diameter. These form when the residual glass viscosity is such that gasses from inorganic decomposition reactions are trapped within the pellet and produce the voids observed.

Figure 9 shows SEM micrographs of material containing 40% waste glass sintered at 1120 ºC. With increasing sintering temperature the material is more effectively sintered but with a bubbled micro-structure containing larger voids than observed in Figure 8c.

4. Discussion

Lightweight aggregates from FA and waste glass have been produced and characterised. Individual lightweight aggregate pellets should ideally have (Cheeseman and Virdi 2005):

- A strong but low density, porous, sintered ceramic core.
- A dense continuous surface layer to inhibit ingress of water that should be pozzolanic to produce a strong aggregate-cement bond in concrete.
- A near-spherical shape to improve fresh concrete properties.

The results obtained show that sintered FA/waste glass pellets can be produced with key properties such as density, water absorption and compressive strength similar or greater than those of commercially available lightweight aggregate products such as Lytag, Aardelite and Optiroc. The properties depend on the sintering temperature, waste glass content and processing conditions. The production of low density, low water absorption and high strength sintered pellets is feasible using relatively simple processing technology involving pelletising and low temperature sintering. In particular, mixes containing 60% FA and 40% waste glass could be sintered over a relatively wide temperature range to produce pellets with appropriate properties for use as lightweight aggregate. The microstructure of optimum 60% FA and 40% waste glass lightweight aggregates contains isolated spherical voids formed by the evolution of gas at temperatures where the glassy phase has low viscosity. The sintering temperature required to form lightweight aggregate from FA and waste glass was between 1080 and 1120ºC. Further work is needed to fully characterise the effect of variations in the chemical composition of FA on sintered properties and the work reported has only investigated FA obtained from the Megalopolis power station in Greece.

The technical feasibility of using lignite coal FA and waste glass in the manufacture of lightweight aggregate has therefore been demonstrated at laboratory scale. However there remain significant barriers to commercial exploitation of the technology. The economic viability is determined by the cost of manufacturing and transporting raw material and products. The fact that FA and glass are readily available waste materials in Greece means that the raw material costs are likely to be low. Subsequent processing involving pelletising and sintering in a rotary kiln is expected to be the same as for other commercially available lightweight aggregates that are typically manufactured from shales, clays and slates. Therefore the associated costs are expected to be similar. However, the manufacture of lightweight aggregate in Greece would avoid the costs and environmental impacts associated with transporting imported materials, and this is likely to make FA/glass lightweight aggregate cost competitive for use in this region.

Other important factors include the ability of the lightweight aggregate formed to meet relevant product standards and codes of practice. There is also likely to be competition from imported lightweight aggregates manufactured from other materials. The large amounts of FA and waste glass available in Greece mean that a FA/glass product has potential to satisfy a significant part of the lightweight aggregate market. The FA/glass lightweight aggregate may also have to compete with natural pumice, although use of this is likely to decline significantly in the future. The research has demonstrated a potential reuse application for lignite coal FA and waste glass and a full technical and economic feasibility study of commercial scale lightweight aggregate production in Greece from FA and waste glass is now required.

5. Conclusions

- The major crystalline phases present in FA from the Megalopolis Power Station are quartz (SiO_2), gehlenite ($Ca_2Al_2SiO_7$), anorthite ($Ca_2Al_2SiO_8$), anhydrite ($CaSO_4$), hematite (Fe_2O_3) and calcite ($CaCO_3$).
- Lightweight aggregates can be produced at relatively low firing temperatures by blending and pelletising FA and waste glass.
- The glass lowers the firing temperature and improves the physical and mechanical properties of the sintered lightweight aggregate product.
- Sintering 60% FA -40% waste glass blends at 1120 ºC produced pellets with a mean density of 1.35g/cm^3, water absorption of ~16% and crushing strength of 7.3 MPa. Lytag had a mean density of 1.34 g/cm^3, water absorption of ~17 and crushing strength of 6.3 MPa.
- The center and the edge of sintered pellets had different micro-structures with different crystalline phases present. The major crystalline phases in the centre were wollastonite and diopside, whereas at the edge the major crystalline phases were moissanite, albite and hematite.
- There is significant potential to manufacture high quality lightweight aggregate from Megalopolis FA and glass that are currently disposed of as wastes to landfill or mines. This could replace natural pumice that is currently used as lightweight aggregate in Greece or other types of imported manufactured lightweight aggregates.

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Figure(s)

Table 1. Waste glass generation and recycling in Greece, 1997-2005

Year	Waste glass generated	Material recycling	Recycling rate
1997	137,500	34,000	25.0
1998	159,500	34,000	21.0
1999	178,500	34,000	19.0
2000	180,000	43,000	24.0
2001	180,000	44,000	24.0
2002	185,000	45,000	24.0
2003	180,000	45,000	25.0
2004	183,000	64,000	35.0
2005	207,000	50,000	24.2

Table 2. Chemical Composition of Greek fly ashes and waste glass

CHEMICAL COMPOSITION	SES AG.DIMITRIOU	SES KARDIAS	SES PTOLEMAIDAS	SES AMINTAIOU	SES MEGALOPOLIS	WASTE GLASS
SiO_2	33-42	26-34	28-41	31-38	47-52	70-74
Al_2O_3	9-12	12-17	13-19	17-21	12-22	1-3
CaO	29-38	33-41	26-39	27-35	5-15	5-11
Fe_2O_3	5-6	4.5-5.5	4-7	2.5-4.0	5-10	
TiO_2	0.7-0.9	0.2-0.5	0.2-0.5	0.7-1.2	N.A.	
MgO	4-6	2-6	2-4	3-5	1.5-3.0	1-3
K_2O	0.8-1.0	0.5-1.0	1.2-1.5	0.6-1.2	1.5-3.0	
Na_2O	0.5-1.5	0.3-0.6	0.3-0.8	0.5-0.7	0.3-0.7	12-16
SO_3	4-9	6-8	6-8	4-9	3-5	

(N.A. - not analysed)

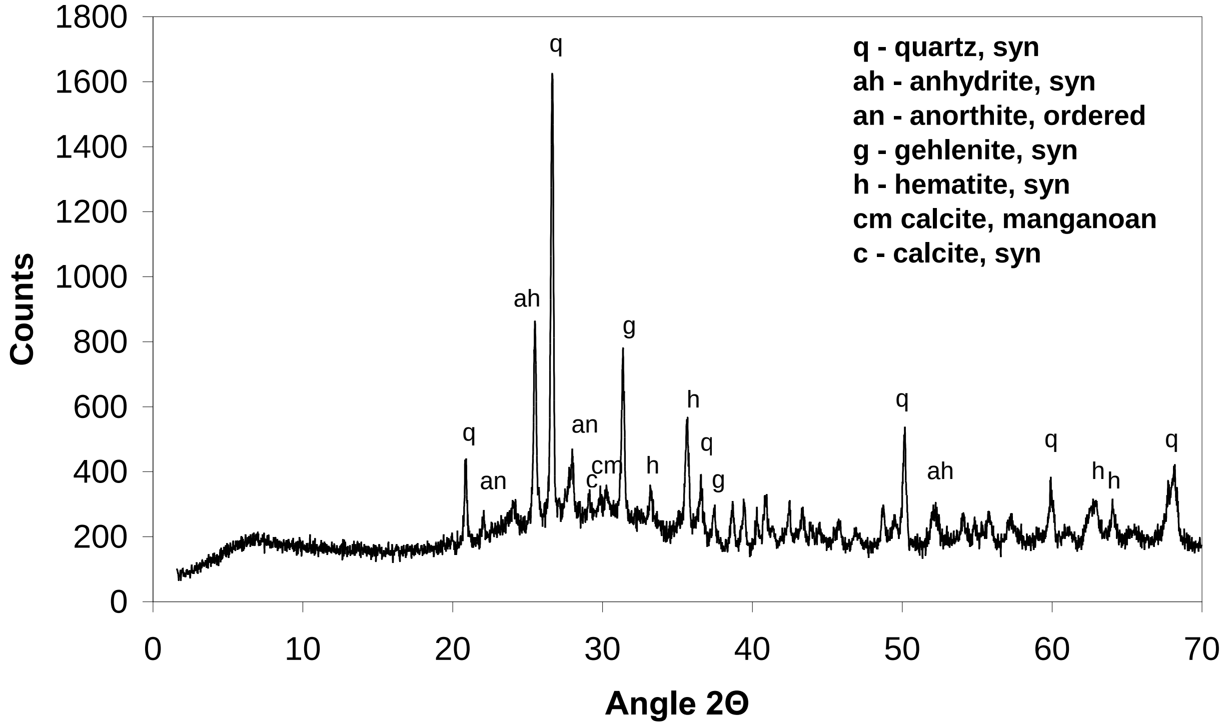
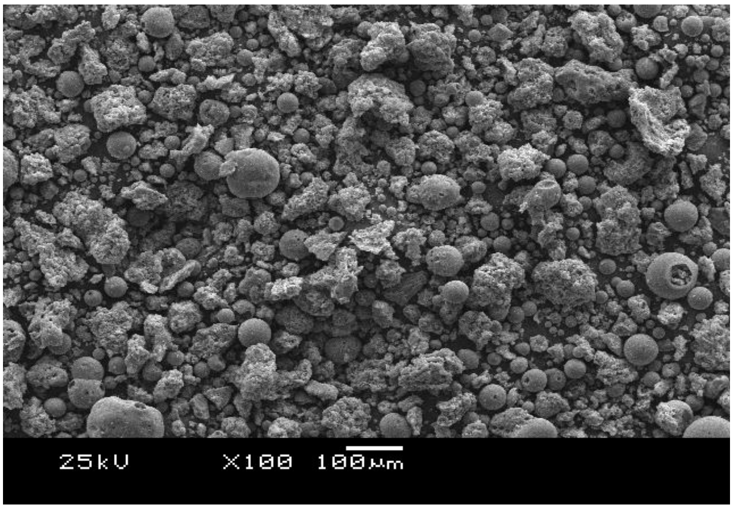
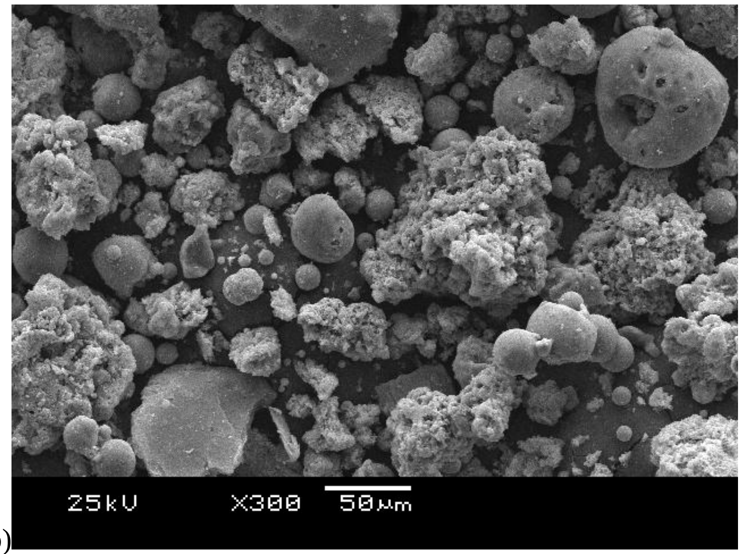


Fig. 1. X-ray diffractogram of <150μm Megalopolis fly ash



(a)



(b)

Fig. 2. Morphology of Megalopolis fly ash (a) x100 (b) x300

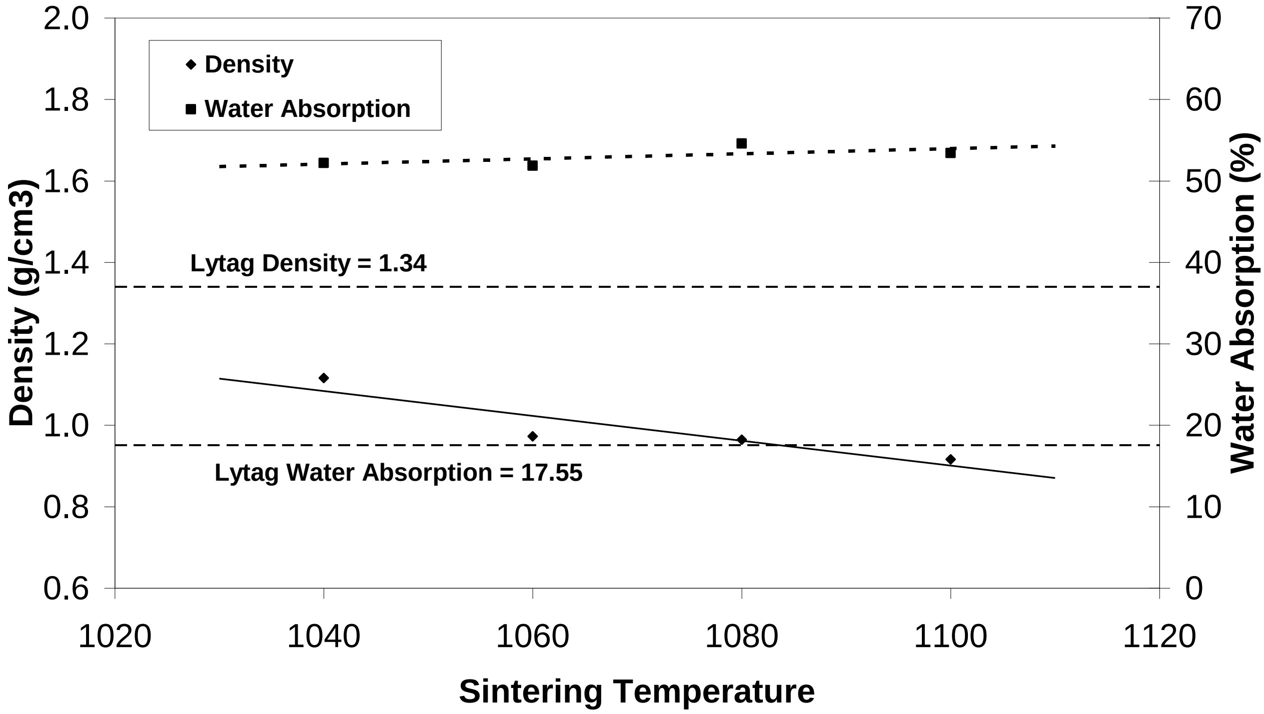


Fig. 3. Effect of sintering temperature on the density and water absorption of sintered Megalopolis fly ash

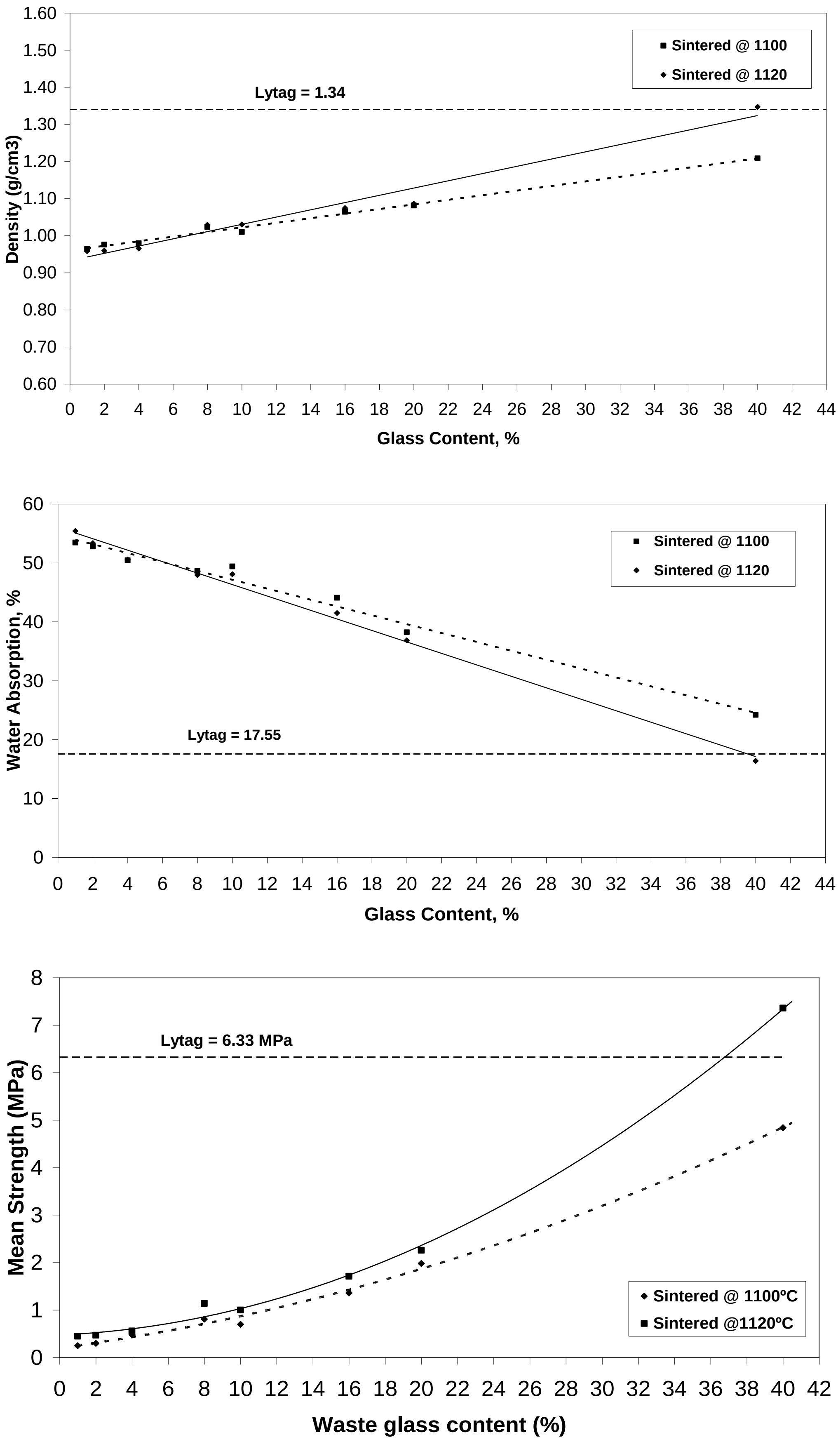


Fig. 4. Effect of glass content on the density, water adsorption and strength of fly ash pellets sintered at 1100 and 1120ºC.

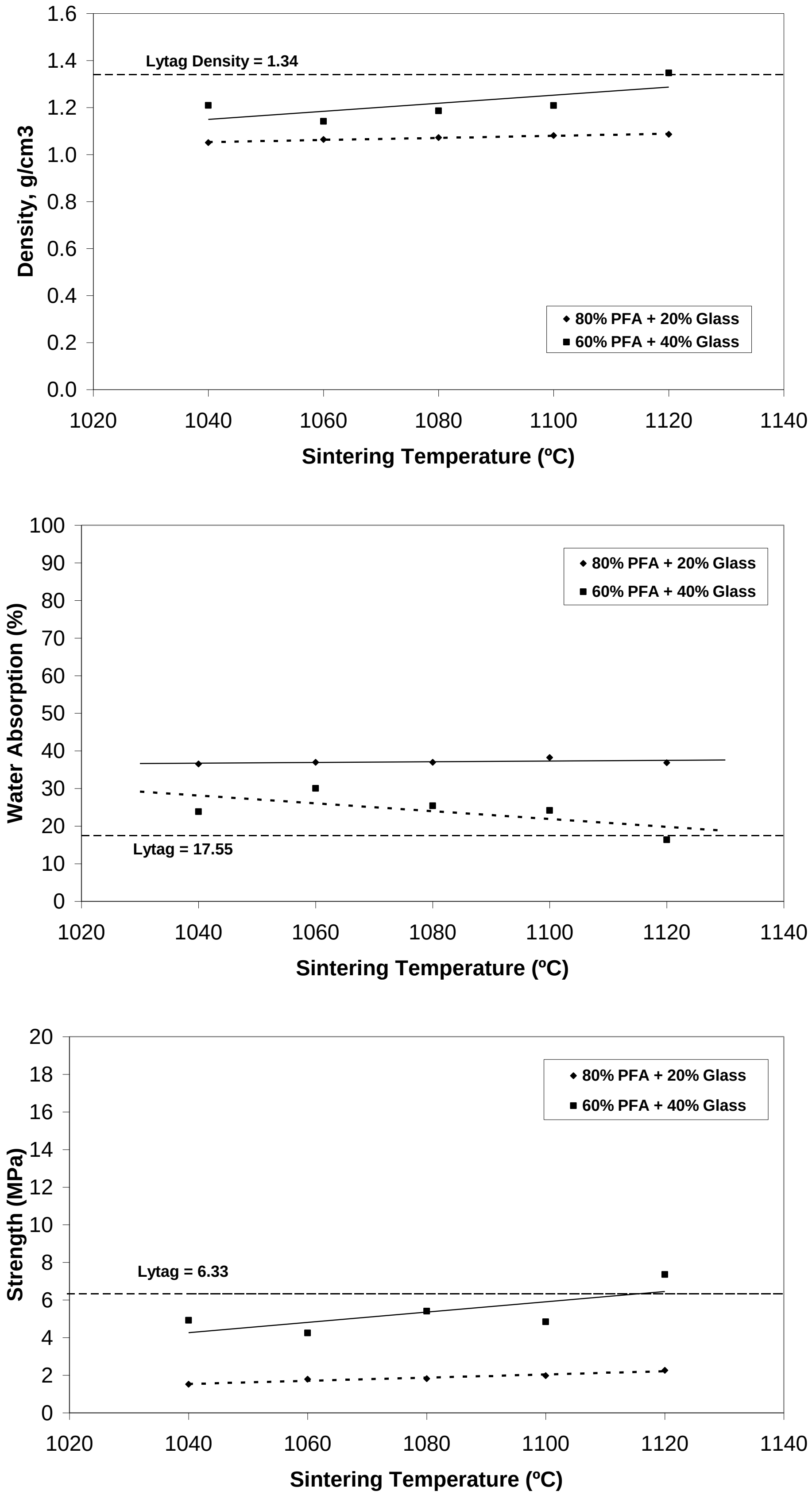


Fig. 5. Effect of sintering temperature on the density water adsorption and strength of 80/20 and 60/40 fly ash/glass mixes.

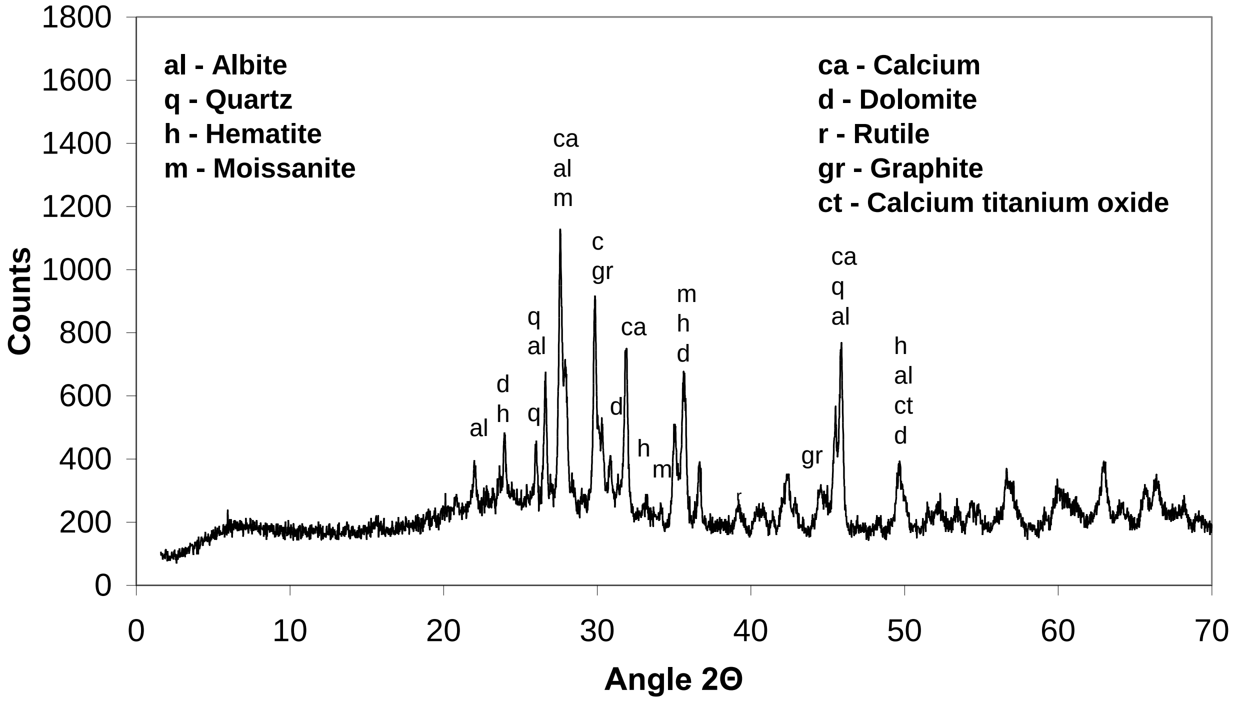


Fig. 6. Outer part of sintered material with 60% fly ash and 40% glass at 1120^{o}C

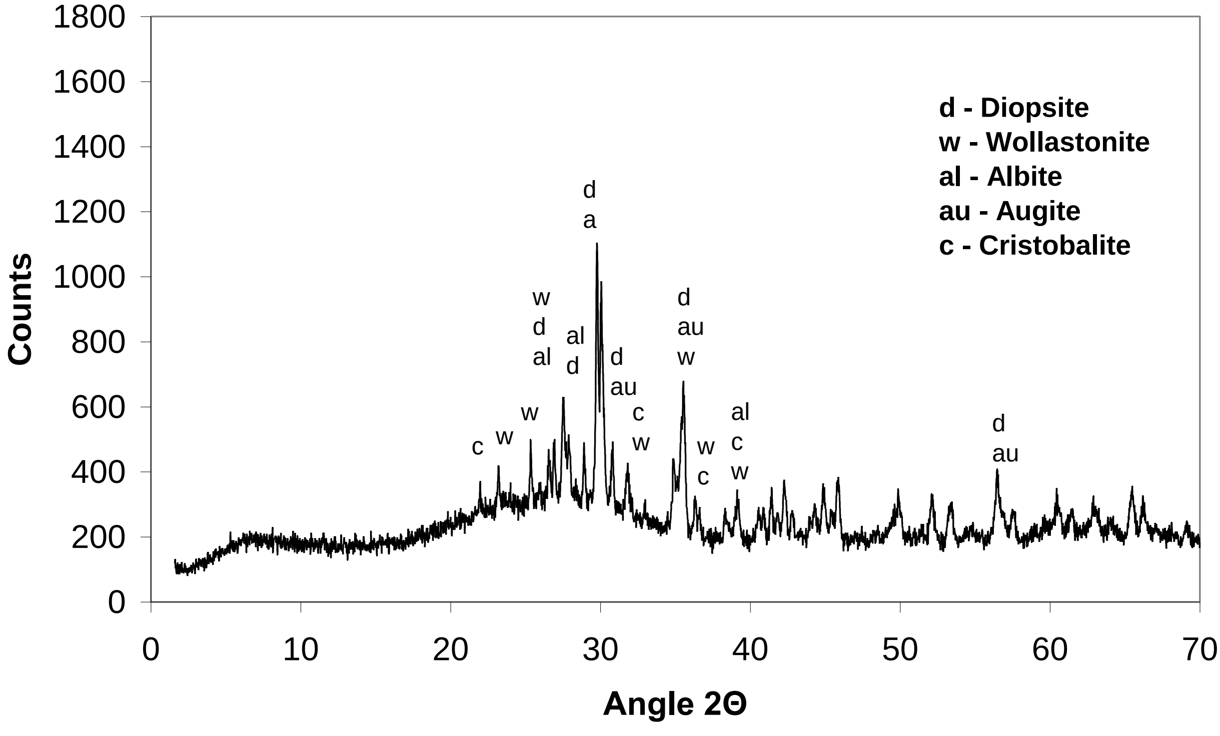
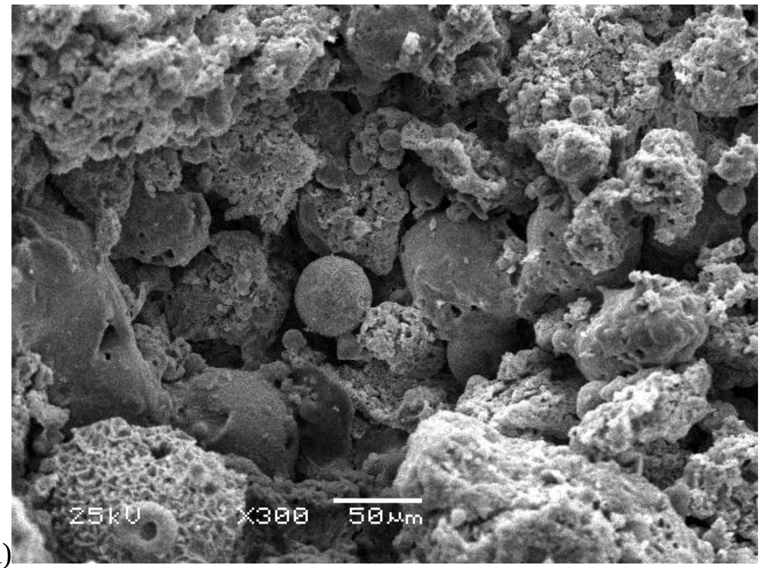
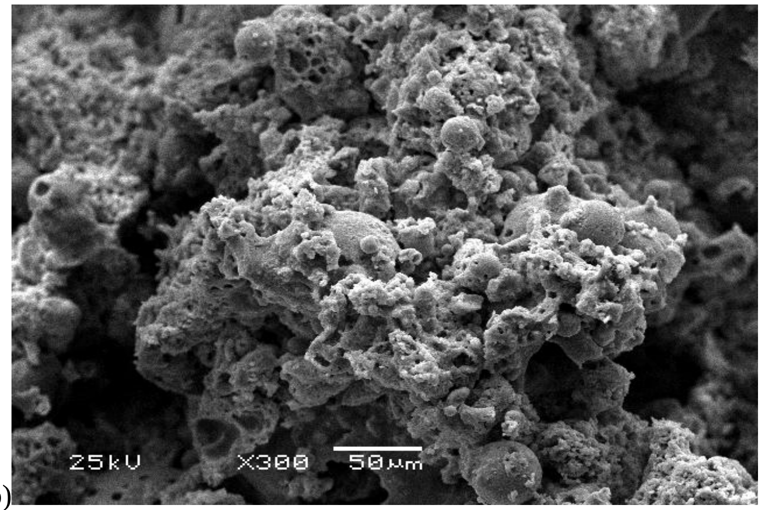


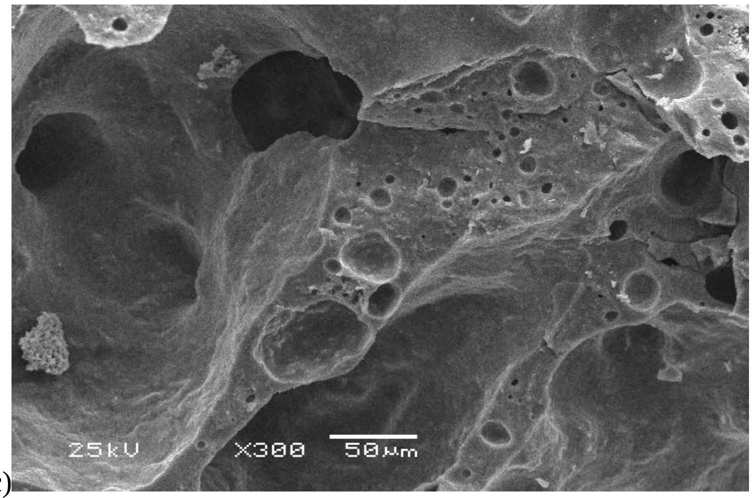
Fig. 7. Inner part of sintered material with 60% fly ash and 40% glass at 1120^{o}C



(a)

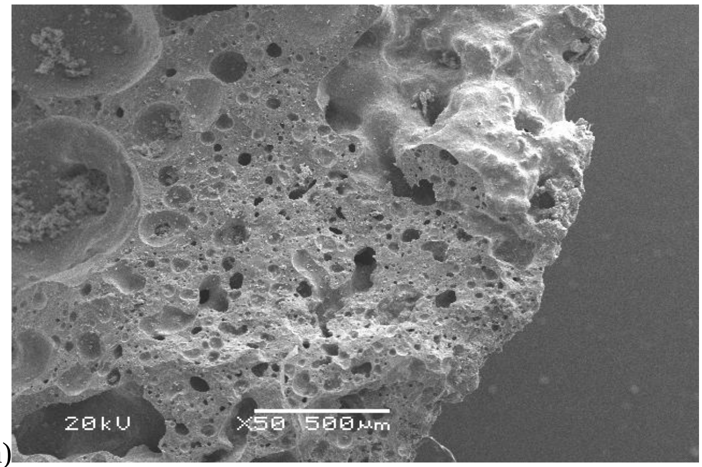


(b)

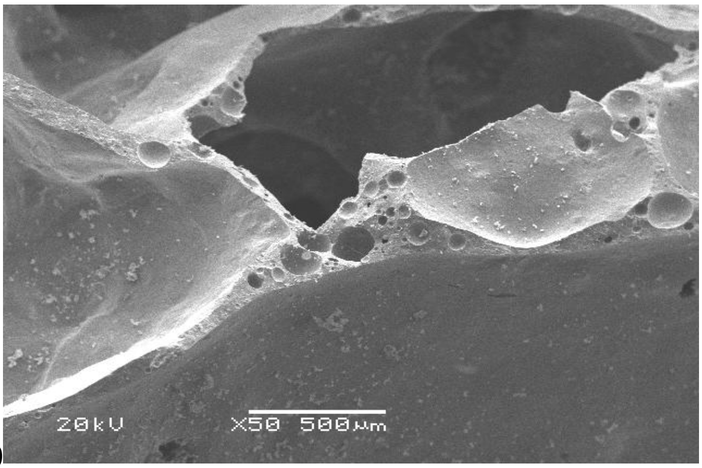


(c)

Fig. 8. Fracture surfaces of LWA pellets sintered at 1100°C containing: (a) 100% FA, (b) 80% FA and 20% waste glass and (c) 60% FA and 40% waste glass



(a)



(b)

Fig. 9. SEM images of samples containing 60% FA and 40% Glass sintered at 1120^{o}C: (a) Edge of sample, (b) Centre of sample