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Characteristics of lightweight fly ash aggregates produced with different binders and heat treatments

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ABSTRACT

This study investigated characteristics of lightweight fly ash aggregates with various binders produced by different heat treatments. Different amounts of binders were mixed with fly ash and put into the pelletization disc. Bentonite and glass powder were used as binders at 0%, 5% and 10%, individually, and in combination at 7.5% each, by weight of fly ash. The pellets thus produced were then sintered at temperatures of 1100, 1150 and 1200 °C. Specific gravity, water absorption, porosity and crushing strength of these aggregates were determined. The properties and microstructure of aggregates were modified by different sintering agents and heat treatments. The strength and specific gravity of all aggregates decreased with increasing the binder content (disregarding binder type) at the highest temperature. Aggregates with low water absorption and high strength and specific gravity can be attributed to the dense structure with discontinuous and small size pores. Increasing binder content reduced water absorption at all temperatures.

1. Introduction

Approximately 15 million tonnes of fly ash are generated annually from thermal coal-fired power plants in Turkey and only a very small proportion of it is used in the construction industry and for various other purposes. As large quantities of the fly ash remain unutilized in most countries of the world, the manufacture of lightweight fly ash aggregates is an appropriate step to greatly increase its use. Recently, some attempts have been made to use fly ash for the production of lightweight aggregates by applying cold bonding, autoclaving or sintering procedures for hardening of the fly ash pellets.

Cold bonding, being a serious low energy alternative to sintering, however, results in lightweight fly ash aggregates with generally inferior properties compared to sintered aggregates [1]. Studies have been made to explore the influences of various hardening methods [2] and the physical and chemical characteristics of the fly ash [3] on the properties of cold bonded fly ash aggregates. Investigations were also reported on the influence of cold bonded fly ash aggregate content [4,5] and the surface treatment of the lightweight fly ash aggregates [6] on the strength, stiffness, absorption as well as shrinkage and creep characteristics of concrete.

Physical properties and strength of artificial lightweight sintered aggregates produced from various industrial by products were previously investigated [7–10]. One advantage of using pulverized coal fly ash compared to other types of ashes in the production of sintered lightweight aggregate is that it promotes fuel efficiency because burning of the carbon in fly ash supplements the heat needed to evaporate the moisture in the pellets and to bring them to the sintering temperature [11]. The suitability of fly ash and different binders for the pelletization and sintering process, on the other hand, is difficult to predict because many physico-chemical factors are involved. Ramamurthy and Harikrishnan [12] investigated the effect of binders on the properties of fly ash lightweight aggregates sintered at 1100 °C and they have observed that the specific gravity, water absorption and strength of the aggregates depended on the type (cement, lime and bentonite) and amount (0%, 10%, 20% and 30%) of the binder which did not alter the chemical composition but the microstructure of the aggregate. Test results have shown that bentonite performed better than cement and lime by significantly improving the 10% fines value and reducing the water absorption of the aggregates [12]. Manikandan and Ramamurthy [13] discussed the relative performance of bentonite with two different swelling characteristics on the pelletization of fly ash and reached to the conclusion that the maximum 10% fines value and minimum water absorption result at the maximum level of factors such as angle and speed of the disk, duration of pelletization, moisture content and bentonite content. After testing few organic and inorganic binders quantitatively for the strength of fly ash pellets sintered at temperatures up to 800 °C Yoo and Jo [14] observed that alkali and acidic materials enabled to produce strong pellets whereas lime and cement acted more efficiently at low temperatures. The manufacturing processes

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and the pore structure of sintered fly ash lightweight aggregate (Lytag) were investigated [15,16] and the microstructure was modified by heat and polymer treatments to obtain aggregates different in their strength, absorption and pozzolanic activity [17]. Wasserman and Bentur [17] have also shown that the strength of the concrete could not be accounted for by the strength of the aggregates only and it was suggested that the absorption and pozzolanic activity of the aggregates could have an influence on the strength developed. Many countries are producing fly ash lightweight aggregates commercially under different trade names, such as Terlite, Lytag, Waylite, etc. [18]. Commercially produced lightweight aggregates (Lytag) were used in concrete by many researchers [19–24] to investigate the strength, stiffness and durability of concrete.

Results of these investigations on fly ash lightweight aggregate indicate that one can expect not only the protection of environment through recycling of waste resources but also the reduction in weight and enhancement of some properties of concretes. Although considerable number of research on the lightweight aggregate production from fly ash and other waste materials are available in the literature limited number of papers has been published on the use of bentonite and glass powder separately and together for improving pelletizing and sintering behavior as well as the properties of the lightweight aggregates. The main objective of this investigation was to study the physical and mechanical properties of lightweight fly ash aggregates produced with different binders (bentonite and/or glass powder) at different sintering temperatures Glass powder was utilized as an alternative to bentonite, which was mostly used in previous studies, for the purpose of investigating the effects of differences in chemical composition, mineralogy and crystallinity as well as physical characteristics of the binders on the pelletization process and sintering behavior of the fly ash–binder combinations as reflected in the properties of the lightweight aggregates produced. Therefore, specific gravity, porosity (water absorption for open porosity, mercury intrusion porosimetry (MIP) for total and closed porosity and for critical pore diameter and pore size distribution) and crushing strength of the lightweight fly ash aggregates with six different fly ash-binder compositions, sintered at three different temperatures (1100, 1150 and 1200 °C) were determined.

2. Materials and methodology

2.1. Properties of materials used in the study

Chemical compositions of fly ash, bentonite and glass powder are shown in Table 1. Quantachrome-NOVA 2200e BET (Brunauer Emmett–Teller) method was used to determine the specific surface area of the fly ash from N_2 adsorption isotherms. The BET method is commonly used on powders to obtain reliable surface area measurements (<10% error). Fly ash used in this study conforms to the requirements of Class F as specified by ASTM C 618 which is shown in Table 2. Particle size distributions of the powder materials used in lightweight aggregate production are illustrated in Fig. 1. Mastersizer was used to determine the grading curves of these materials by laser diffraction. It can be seen that fly ash has the coarsest and bentonite has the finest particle size distribution whereas the particle size distribution of glass powder is close to that of fly ash. Bentonite contains 33% of particles smaller than 10 μm, whereas glass powder and fly ash contain 21% and 17% of particles smaller than 10 μm, respectively.

Table 1
Chemical composition of fly ash, bentonite and glass powder (wt.%).

Oxide (%)	Fly ash	Bentonite	Glass powder
SiO_2	59.00	57.84	70.62
Al_2O_3	19.58	13.77	1.38
Fe_2O_3	7.23	6.14	0.82
CaO	0.54	3.75	8.75
MgO	4.64	3.04	3.54
SO_3	0.69	1.34	1.85
Na_2O	0.48	2.80	10.85
K_2O	5.95	2.80	1.53
Cl^-	0.0114	0.0035	0.0025
Loss on ignition	0.49	8.40	0.12
$CaCO_3 + MgCO_3$	–	6.50	0.75
Specific surface area (m^2/kg)	288	–	–
Specific Gravity	2.06	2.40	2.50

2.2. Preparation of lightweight aggregates

Harikrishnan and Ramamurthy have shown that the pelletization process parameters such as the speed and the angle of the pelletizer disk, moisture content and the duration of the pelletization strongly influence the properties of the fly ash aggregates produced [25]. A pelletizer disc of 400 mm diameter and 150 mm depth was used for manufacturing the fly ash pellets, and the disc was operated at an angle of 43° and a speed of 45 rpm based on the findings of a previous study [26]. Binders like bentonite (B) and glass powder (G) were added as sintering agents at 5% and 10%, individually, by weight of fly ash (FA). Fly ash without a binder and fly ash with the combination of these two binders (B7.5% + G7.5%) were also used for making pellets. Water was sprayed onto the powder mixture in the pelletizer at 22–25% by weight of the total material in order to form the spherical pellets. Less water was required to wet the particles when bentonite was included in the mixture. Addition of glass powder, however, did not alter the water content.

Irregular particles were obtained when 15% bentonite was added to fly ash. An increase in dosage of bentonite beyond this level resulted in sticking of pellets to each other leading to formation of muddy balls, which was attributed to the high plasticity index and higher expansive nature of three-layer montmorillonite structure of the bentonite [27]. Ramamurthy and Harikrishnan [12], on the other hand, produced aggregates containing bentonite content up to 30% with good ballability. The differences in the speed and

Table 2
Chemical properties of fly ash and specification requirements.

Properties (%)	Fly ash used	ASTM C 618 (Class-F)
$SiO_2 + Al_2O_3 + Fe_2O_3$	85.81	70 (min)
MgO	4.64	5 (max)
CaO	0.54	<10
SO_3	0.69	5 (max)
Loss on ignition	0.49	6 (max)

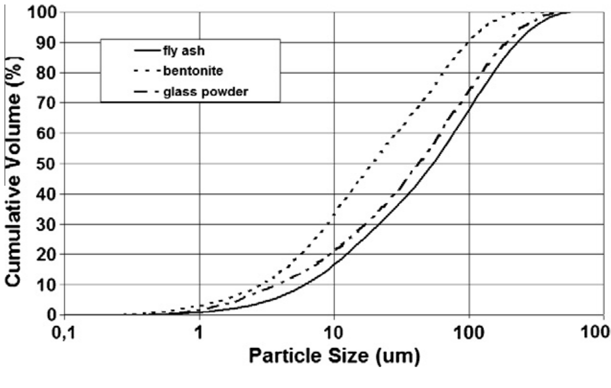

Fig. 1. Particle size distribution of fly ash, bentonite and glass powder.

the angle of the pelletizer, moisture content and properties of the fly ash might have caused this discrepancy between this study and the one reported in [12]. Also, pelletizing fly ash with 15% glass powder resulted in pellets with larger diameter. For these reasons, in this study, the upper limit for both bentonite and glass powder content was selected as 10%. In addition, insufficient water led to difficulty in pelletization and surplus water caused bigger and irregular particles, as also confirmed by Chiou [28].

The pelletizer disc was rotated until firm spherical pellets have been formed which took approximately 20 min. The pelletizing time was kept constant for each mixture. After pelletization, the pellets were first dried in an oven at 110 °C to prevent cracking and exploding of the wet pellets during heating. Pretests have shown that the pellets had loosely bound fly ash particles and weak matrix after sintering at temperatures below 1100 °C whereas full densification of only fly ash aggregates and bloating of fly ash aggregates with binders were observed at 1200 °C. Therefore, it was decided to sinter the pellets at 1100, 1150 and 1200 °C temperatures. The average heating rate in the kiln was 6.7 °C/min. The sintering time at maximum temperature was one hour and kept constant for all mixtures. After sintering was over, the kiln was shut and the aggregates were left in the furnace to cool in a slow rate. The heat increase profile in the furnace is shown in Fig. 2. The schematic illustration of the aggregate production is presented in Fig. 3.

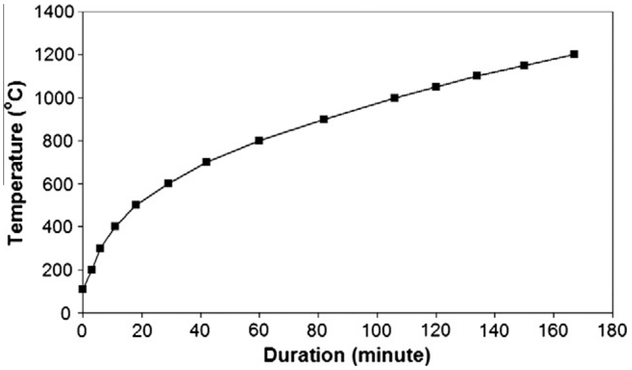

Fig. 2. Heat increase profile in the furnace.

Notation (designation) for the aggregates produced is such that the letters OFA, B, G and BG indicate the materials used in the preparation of pellets as only fly ash, bentonite added, glass powder added and both bentonite and glass powder added, respectively. The preceding number represents the weight ratio (% by weight) of the added binders and the following number is for the sintering temperature.

2.3. Tests and analyses performed on aggregates

2.3.1. Crushing strength

In this study, strength of the lightweight fly ash aggregates was determined by measuring the crushing strength of the individual aggregate pellets by loading them diametrically. The individual aggregate particles were placed between two parallel plates and were crushed by loading diametrically between these parallel plates. The crushing force was measured by a 4.5 kN capacity load-ring. Stress analysis shows that when a spherical particle is compressed on two diametrically opposed points the crushing strength σ is given by:

$$\sigma = \frac{2.8 * P}{\pi * X^2} \tag{1}$$

where X is the distance between loading points and P is the fracture load [8,9,29,30]. Minimum 20 pellets, each with approximately 10 mm diameter were tested to calculate the average crushing strength for each aggregate type.

2.3.2. Specific gravity

Oven dry specific gravity of lightweight aggregates were determined according to ASTM C 127 from measurements of masses of oven dry sample in air, saturated surface dry sample in air and saturated sample in water.

2.3.3. Porosity

Pore structure and open and closed porosity of the lightweight fly ash pellets were investigated.

2.3.3.1. Water absorption of aggregates. Open porosity of lightweight fly ash aggregates were estimated according to ASTM C 127 by calculating the weight percentage water absorption of the

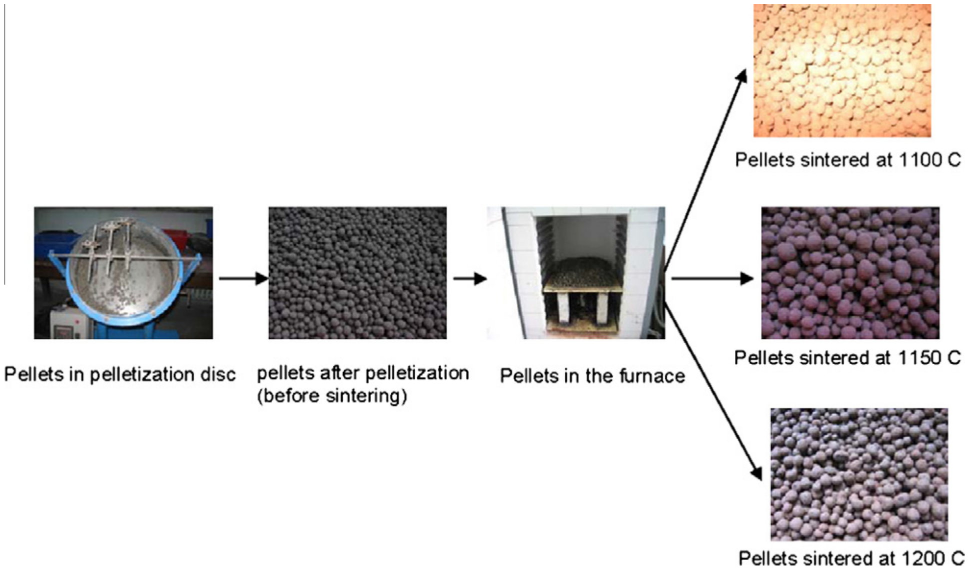

Fig. 3. Production of lightweight fly ash aggregates.

aggregates from measurements of masses of oven dry and saturated surface dry samples in air.

2.3.3.2. Mercury intrusion of aggregates. Pore structure (total porosity and pore size distribution) of the lightweight fly ash aggregates was estimated by mercury intrusion porosimetry with a pressure applied up to 220 MPa. Mercury intrusion porosimetry (MIP) is a better measure of closed porosity of aggregates. Quantachrome-PoreMaster Mercury porosimetry was used in this study to determine the total porosity of the lightweight fly ash aggregates. The porosity and the critical pore diameter were determined from the pore size distribution curves for each aggregate. The method for the preparation of the samples for the MIP test can have impact on the porosity values; for example, mechanical chipping and subsequent drying can lead to microcracks in the samples. Therefore, individual aggregate particles with diameter smaller than 8 mm were used for MIP tests.

Assuming that the pores have cylindrical shape, the diameters of the pores were computed according to Eq. (2) using the pressure and intruded quantity of mercury [31–34],

$$d = -4 * \gamma * \cos\theta / P \qquad (2)$$

where d is the pore width, P is the net pressure across the mercury meniscus at the time of the cumulative intrusion measurement, γ is the surface tension, and θ is the contact angle between mercury and the pore wall. In this study, the values for γ and θ were 0.480 N/m and 141.3°, respectively.

3. Test results and discussion

3.1. Effects of temperature and type and content of the binder on specific gravity

Effects of the sintering temperature, type and content of the binder on oven dry specific gravity of the fly ash aggregates are exhibited in Table 3. Specific gravity values increased continuously with the increasing sintering temperature in the case of OFA aggregates. Obtaining denser structures and higher specific gravities can be attributed to the excessive glass formation at higher temperatures as also observed by Al-Bahar and Bogahawatta [35]. It is also noted that the aggregates showed substantial changes in specific gravities with increasing binder concentration. The highest and the lowest specific gravity values for OFA aggregates were obtained by sintering at 1200 and 1100 °C, respectively; whereas the aggregates with a binder sintered at 1150 and 1200 °C mostly gave the highest and lowest specific gravities, respectively.

Addition of binders (disregarding binder type) increased the specific gravity values at 1100 and 1150 °C due to the fact that some of the binder particles were retained unreacted at lower temperatures. These retained particles closed and insulated the interstitial voids, leading to low water absorption and high specific gravity. In other words, addition of binders increased the specific gravity at lower sintering temperatures due to the incomplete reaction resulting in insufficient gas evolution to form pores within the fly ash pellets. As can be seen from Table 1, specific gravities of glass powder and bentonite are higher than the specific gravity of fly ash. Thus, addition of these binders to fly ash naturally increased the pellet specific gravity at low sintering temperatures, in the absence of sufficient heat for bloating. At 1100 and 1150 °C sintering temperatures, increasing bentonite content from 5% to 10% increased the specific gravity from 1.75 to 1.77 and from 1.90 to 1.93, respectively, while with the same increase of glass powder content, specific gravity decreased from 1.81 to 1.77 and from 1.93 to 1.85, respectively. Nevertheless, all of the aggregate types produced had specific gravities below 2.0 to be identified as lightweight.

Table 3
Oven dry specific gravity, water absorption and crushing strength of lightweight fly ash aggregates.

Temp. (°C)	Property	OFA	5B	10B	5G	10G	7.5BG
1100	OD. spec. grav.	1.51	1.75	1.77	1.81	1.77	1.78
	Water abs. (%)	18.4	8.4	6.9	11.6	8.2	4.5
	Crushing str. (MPa)	5.1	13.1	13.7	9.7	11.5	14.0
1150	OD. spec. grav.	1.74	1.90	1.93	1.93	1.85	1.73
	Water abs. (%)	10.4	2.3	0.8	3.4	2.6	0.9
	Crushing str. (MPa)	8.6	20.4	23.1	15.6	14.7	14.2
1200	OD. spec. grav.	1.92	1.62	1.56	1.66	1.59	1.34
	Water abs. (%)	2.2	1	0.7	1.1	0.7	0.8
	Crushing str. (MPa)	19.3	13.4	12.0	11.8	9.6	7.1

Test results also indicated that the use of glass powder and bentonite led to a significant reduction in specific gravity of the resulting aggregate when sintered at 1200 °C. The effect was more prominent in the aggregates containing higher content of binders. When 7.5% glass powder and 7.5% bentonite were added as binders to fly ash, a greater bloating effect was noticed as was evident from the relatively low values of specific gravity of the aggregate produced. This is due to the increase in the quantity of gases evolved during sintering at high temperature. The decrease in density at high temperatures which has also been reported by other researchers [36–41] was attributed to the internal sintering due to the calorification and combustion of organic materials and the accelerated expansion due to gas generation. Adell et al. [40] reported that an initial marked contraction followed by an expansion stage was observed with increasing sintering temperature and all samples started to expand at temperatures above 1200 °C. This reduction in density was also attributed to internal pressure caused by the formation of CO_2 (and possibly CO) upon combustion of the carbon [42]. On the other hand, it was demonstrated that carbon was oxidised and released from the samples at temperatures below 850 °C under conventional, slow sintering conditions [43]. However, Huang et al. [44] stated that the surface of the sample was not completely vitrified when gas was produced, making it possible for gas to escape. In the present study, it was observed that as the surface of the pellets became more sticky by glassy materials more of the gas produced with the increase in internal temperature had been wrapped up in the particles, which made the aggregates lighter.

Glass powder and bentonite contained high concentrations of calcium containing minerals, such as calcium oxides and carbonates as well as sodium and potassium oxides and sulfates (Table 1). These minerals act as fluxes which lower the melting point, causing sintering and densification at lower temperatures. Fly ash contained less of low melting point fluxes and more of high melting point minerals compared to glass powder and bentonite. Density of the aggregates with binders increased with temperature from 1100 to 1150 °C because in this temperature range, as the aggregate sintered, the fluxing agents in the glass powder and bentonite melted to form a liquid phase that filled voids between particles through capillary action. Density increased as voids were filled and as the sintering temperature increased, more of the compounds in the binders melted, substantially eliminating all the pores and forming a larger glassy, crystalline solid matrix. At the temperature of maximum densification, essentially all of the void space in the aggregate were filled. Density decreased with increase in temperature from 1150 to 1200 °C, on the other hand, because further temperature increase resulted in aggregate melting and bloating.

Bloating was caused by the entrapment of gases in the melted liquid phase, resulting from volatization of certain components of the aggregate. The entrapped gases formed the pores. The proper viscosity could be determined from the chemical compositions of the raw materials used [37,39,40]. Generally, total CaO and MgO contents account for a sudden change in the viscosity of the raw materials at high temperatures. Fly ash did not cause a sudden change in the viscosity even at high temperatures because its total CaO and MgO content was 5.18%, compared to 12.29% and 6.79% for glass powder and bentonite, respectively. In general, when the $Al_2O_3 + SiO_2$ content is high, the melting point and viscosity tend to be high. Thus, it is expected that the melting point of the fly ash would be higher than those of bentonite and glass powder.

3.2. Effects of temperature and type and content of the binder on porosity

Variation in water absorption of the lightweight aggregates with sintering temperature and type and content of the binder is shown in Table 3. Water absorption of all aggregate types declined with the increase in sintering temperature. OFA aggregates showed linear decrease, however aggregates with binders exhibited a significant decrease from 1100 to 1150 °C, and then the drop was slight up to 1200 °C. Sintering OFA aggregates at 1100 °C led to loosely bounded fly ash particles with extensive volume and connectivity of the internal and surface pores resulting in the high degree of water absorption. A decrease was observed with the increase of binder content regardless of binder type as also observed by Ramamurthy and Harikrishnan [12]. The reduction was larger in bentonite aggregates than in glass powder aggregates at 1100 and 1150 °C and the same at 1200 °C. The disconnected porosity close to the surface which occured at 1200 °C led to further decrease in water absorption which was also reported [28,37] to be due to the formation of glassy texture on the surface of the aggregates at higher temperatures. Water absorption of OFA1200 aggregate was close to the aggregates with binders sintered at 1150 °C indicating the efficiency of the use of binders.

Results of total porosity measurements by mercury porosimetry are presented in Table 4 and Fig. 4 to represent the pore size distribution of the aggregates sintered at different temperatures. Such figures illustrate the variation of cumulative pore volume obtained from the intruded quantity of mercury under increasing pressure with the size of the pores obtained from Eq. (2) and help to produce the data in Table 4. In all cases, the total porosity measured by the mercury porosimetry were greater than the water saturation of the aggregates due to the presence of a relatively large volume of closed pores which could be penetrated by mercury only after the rupture of pore walls under the high mercury pressure. It is revealed that increase in the temperature should generally result in greater fusion and sintering leading to reduction in open porosity [17]. However this reduction may not be as high as expected due to increase in the volume of the solid which causes reduction in density. In this study, a lower density aggregate with a microstructure of high porosity within a matrix was targetted and thus bentonite and glass powder were introduced into fly ash as binders. The binders burned off during sintering, increasing the porosity of the resultant aggregates and decreasing their densities. Thus, lightweight aggregates with specific gravities as low as 1.34 could be produced by adding binders. Comparing the density and water absorption data clearly indicated a change in the nature of the porosity as the sintering temperature and the binder content increased which may be explained in terms of the expansion and bloating taking place due to the softening of the glassy phase and gas evolving decomposition reactions occurring in a low viscosity glassy matrix.

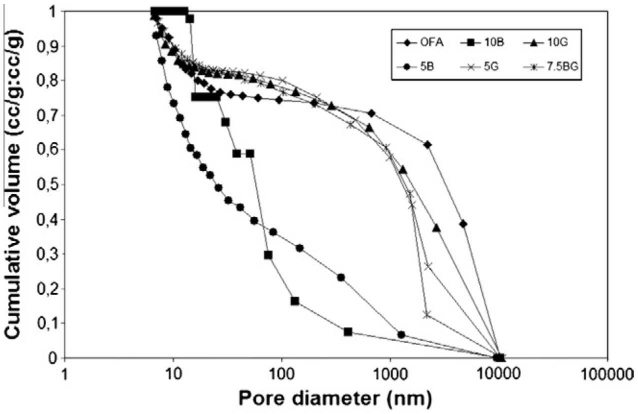

Fig. 4. Pore size distribution of aggregates sintered at 1100 °C.

Table 4
Results of MIP tests.

Aggregates	Pore diameter range (nm)		Critical pore diameter (nm)	Total porosity (%)	Closed porosity (%)
	max	min			
OFA1100	10277.72	7.02	26.92	19.1	0.7
5B1100	10277.75	7.28	32.44	15.3	6.9
5G1100	9535.24	7.01	19.90	13.6	2.0
10B1100	10277.74	6.80	38.63	15.1	8.2
10G1100	10551.59	6.70	15.94	11.3	3.1
7.5BG1100	10840.49	7.16	34.93	13.4	8.9
OFA1150	10017.71	7.16	19.62	13.3	2.9
5B1150	10840.49	7.11	19.44	10.1	7.8
5G1150	10277.72	6.66	17.87	7.3	3.9
10B1150	10551.60	6.60	21.47	2.0	1.2
10G1150	10017.69	6.77	12.16	3.9	1.3
7.5BG1150	9311.02	6.60	49.23	2.5	1.6
OFA1200	10840.51	6.82	16.35	6.5	4.3
5B1200	10277.77	6.88	53.38	17.6	16.6
5G1200	10840.49	6.82	21.64	16.9	15.8
10B1200	10840.46	6.82	28.03	8.4	7.7
10G1200	8159.76	7.08	12.49	6.2	5.5
7.5BG1200	10840.49	6.68	74.91	21.8	21.0

There was a linear reduction in total porosity of OFA aggregates as in water absorption. Inclusion of binders reduced the total porosity at 1100 and 1150 °C, but increased at 1200 °C. At each temperature level, total porosity decreased when the content of binders increased except for 7.5BG1200 aggregates. However, this behavior is contradictory with the results of specific gravity and crushing strength at 1200 °C. The reason may be due to the formation of excessive glassy phase for higher binder contents at 1200 °C close to the surface of the particles preventing the penetration of mercury. In this study, 220 MPa mercury pressure applied on aggregates may have been insufficient to rupture the thick and strong pore walls developed under such circumstances.

The closed porosity of OFA, 5B and 5G aggregates increased with the rise in temperature. This increase was remarkable in OFA aggregates sintered at 1150 °C, however, a sharp increase was observed in aggregates with 5% of binder at 1200 °C. The aggregates with higher binder content showed decrease in total porosity at 1150 °C. Total porosity of aggregates with 10% bentonite and glass powder exhibited decreases from 15.1% to 2.0%, and from 11.3% to 3.9%, respectively. In addition, aggregates produced with both glass powder and bentonite showed a drop from 13.4% to 2.5%. These changes in porosity were in consistence with the trend observed in specific gravity and strength. 7.5BG1200 aggregate had the highest closed porosity due to the high amount of binders resulting in occurence of insulated larger pores after sintering at high temperature. While OFA1100 aggregate had much higher water absorption capacity than 5B1200, 5G1200 and 7.5BG1200 aggregates, these aggregates showed similar total porosities, but indicating a more open, water-accessible pore structure for OFA1100 aggregate compared to the others. 10B1200 and

10G1200 aggregates, however, had less total porosity, but almost all of the pores were unconnected and closed.

The minimum and maximum pore diameters were about 7 and 10000 nm, respectively. The critical pore diameter which corresponds to the smallest pore size that creates connected paths through the sample was obtained from the inflection point of the plot of volume versus size of the pores. Generally, the critical pore diameters of aggregates showed paralellism with the total porosity values. The critical pore diameter decreased with sintering temperature in OFA aggregates. However, aggregates with binders showed a decrease up to 1150 °C, and then a rise with further increase in temperature. A relatively high critical pore diameter was obtained for the 7.5BG1200 aggregate. This aggregate with its high binder content greatly expanded at 1200 °C due to bloating which may have resulted in thin pore walls making the aggregate pore structure more vulnerable to high pressures of the mercury in MIP test.

3.3. Effects of temperature and type and content of the binder on crushing strength

Effects of sintering temperature and binder type and content on crushing strength are shown in Table 3. The aggregates with binders exhibited higher strength than OFA aggregates at 1100 and 1150 °C as also reported by Ramamurthy and Harikrishnan [12]. Crushing strength increased slightly by the binder content at these temperatures except for the G1150 aggregates. Variation in crushing strength could not be correlated only with the variation in porosity as also demonstrated by other studies [12,17]. Several interrelated factors such as change in the mineralogical composition, difference in melting points of binders densification effects by sintering, bloating of the aggregate and cracks and internal defects due to thermal stresses may be effective.

Strength of OFA aggregates continuously increased with the sintering temperature whereas aggregates with binders gained the highest strength at 1150 °C. Decrease in strength was probably because of the large pores formed due to bloating at 1200 °C. Volume expansion observed at different sintering temperatures for different materials was claimed to be the cause of the drop in strength of the aggregates [7,8,10,37–39,41,45]. Thus, steady increase in the strength of OFA aggregates with temperature was achieved through densification of fly ash. Expansion of OFA aggregates probably might take place at temperatures higher than 1200 °C, at which density of OFA aggregates was similar to those of aggregates with binders at 1150 °C. Addition of binders modified the densification behavior of fly ash, resulting in a tighter temperature interval between the initial softening, sintering, and melting of the material by changing the chemical composition and mineralogy of the material.

The chemical composition affects the viscosity and the sintering behavior of fly ash as illustrated by SiO_2 acting as a network former and positively affecting the viscosity whereas CaO or Na_2O modifying the silicate network structure and decreasing the viscosity [40]. The principal chemical compositions of fly ash, bentonite and glass powder given in Table 1 met the requirements of expansive clay (SiO_2: 48–70%, Al_2O_3: 8–25% and Flux (sum of Fe_2O_3, CaO, MgO, Na_2O and K_2O): 4.5–31%) as stated by Riley [46]. Two conditions were essential for the bloating of materials: development of a glassy phase over a wide temperature range and evolution of gases from the dissociation of mineral components such as carbonates, oxides, hydrates, and sulfates. Fly ash is richer in glass forming oxides ($SiO_2 + Al_2O_3$) than bentonite and glass powder. The modifying or fluxing oxide contents of fly ash (18.84%) and bentonite (18.53%) are close to each other and lower than that of glass powder (25.49%) which was the main reason for the viscosity showed by the melted materials. The greater possibility of better bloating could be achieved when bentonite and glass powder were mixed with fly ash at adequate proportions. The addition of glass powder increased the flux content and improved the chances of bloating in the aggregates.

Bentonite was more effective in strength rise than glass powder especially at 1150 °C. The strengths of aggregates with glass powder were lower than those of aggregates with bentonite at all temperatures. Glass powder used in this study was coarser than bentonite (Fig. 1), resulting in larger pores during exposure to high temperatures. OFA aggregates became stronger by the temperature rise and showed marked increase between the temperatures 1150 and 1200 °C. Furthermore, as bentonite and/or glass powder were added, binders closed the pores by fusing, causing denser aggregates at 1100 and 1150 °C. At 1150 °C, strength gain was markedly high in aggregates with 5% binders, especially in the case of bentonite, whereas addition of 10% binders did not provide significant increase in strength, even the aggregates with glass powder presented a slight drop.

Among the aggregates produced, 10B1150 aggregate had the highest (23.1 MPa) and OFA1100 aggregate had the lowest (5.1 MPa) strength values. The strengths of aggregates with binders showed the same trend of pellet densities with increasing temperature, increasing to a maximum value and then decreasing, as expected. Bentonite was finer than glass powder and aggregates made from small particle sizes showed a higher strength, which is consistent with observations obtained by other researchers [10,40]. Since smaller particles give a larger total surface area, small ash particles may result in a larger change in the surface area during sintering, and in turn, a larger change of free energy. The densification and thus the development of the strength is thermodynamically more likely to occur for ash with a smaller particle size.

4. Conclusions

Physical and mechanical properties of lightweight fly ash aggregates produced with and without bentonite and/or glass powder binders sintered at 1100, 1150 and 1200 °C temperatures were investigated. Following conclusions can be drawn from the results and findings of the experimental study:

- Pellets had loosely bound fly ash particles and weak matrix by sintering at temperatures below 1100 °C whereas full densification of OFA aggregates and bloating of fly ash aggregates with bentonite and/or glass powder binders were observed by sintering at temperatures at and above 1200 °C. The particles in the OFA aggregates fused at 1150 °C, whereas melting and fusion occured at 1100 °C with the addition of a binder. Addition of binders modified the densification behavior of fly ash, resulting in a tighter temperature range for the initial softening, sintering and melting of the material.
- Specific gravity of OFA aggregate continuously increased with sintering temperature. Addition of binders increased the specific gravity at low sintering temperatures (1100 and 1150 °C) due to higher specific gravity of the binders compared to fly ash as well as the absence of sufficient heat for bloating and gas evolution to produce large pores. The use of binders led to a significant reduction in specific gravity of aggregates sintered at 1200 °C due to expansion and bloating.
- Water absorption of all aggregates declined with the increase in temperature. Relatively high water absorption, low specific gravity and low strength at 1100 °C were due to open and connected porosity of the aggregate particle. Aggregates with binders had lower water absorption at 1150 °C due to discontinuous porosity, denser structure and small pore size while further decrease in water absorption was obtained at 1200 °C owing to the large but closed pores and formation of glassy texture

on the surface of the aggregate particles. Water absorption of OFA1200 aggregate was close to the aggregates with binders at 1150 °C indicating the efficiency of the binders.

- Total porosity continuously decreased with the increase in temperature for OFA aggregates. Inclusion of binders, however, reduced the total porosity at 1100 and 1150 °C, but increased it at 1200 °C. While the critical pore diameter continuously decreased with temperature in aggregates without binder, for the aggregates with binder it decreased up to 1150 °C and then raised with further increase in temperature.
- Strength of aggregates with bentonite and glass powder binders at 1100 and 1150 °C were much higher than OFA aggregate and increased with binder content. Bentonite was more effective in increasing the strength than glass powder especially at 1150 °C. The highest strength for OFA aggregates was obtained at 1200 °C whereas the strength of aggregates with binder at 1200°C were reduced due to bloating effect resulting in large closed pores.

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References

[1] Kockal NU, Ozturan T. Effects of lightweight fly ash aggregate properties on the behavior of lightweight concretes. J Hazard Mater 2010;179(1–3):954–65.

[2] Manikandan R, Ramamurthy K. Effect of curing method on characteristics of cold bonded fly ash aggregates. Cem Concr Compos 2008;30(9):848–53.

[3] Gesoglu M, Ozturan T, Guneysi E. Effect of fly ash properties on characteristics of cold bonded fly ash lightweight aggregates. Construct Build Mater 2007;21(9):1869–78.

[4] Joseph G, Ramamurthy K. Influence of fly ash on strength and sorption characteristics of cold bonded fly ash aggregate concrete. Construct Build Mater 2009;23(5):1862–70.

[5] Gesoglu M, Ozturan T, Guneysi E. Shrinkage cracking of lightweight concrete made with cold bonded fly ash aggregate. Cem Concr Res 2004;34(7):1121–30.

[6] Gesoglu M, Ozturan T, Guneysi E. Effects of cold bonded fly ash aggregate properties on the shrinkage cracking of lightweight concretes. Cem Concr Compos 2006;28(7):598–605.

[7] Cheeseman CR, Virdi GS. Properties and microstructure of lightweight aggregate produced from sintered sewage sludge ash. Resource Conserv Recycling 2005;45(1):18–30.

[8] Cheeseman CR, Makinde A, Bethanis S. Properties of lightweight aggregate produced by rapid sintering of incinerator bottom ash. Resource Conserv Recycling 2005;43(2):147–62.

[9] Mangialardi T. Sintering of msw fly ash for reuse as a concrete aggregate. J Hazard Mater 2001;B87:225–39.

[10] Wang KS, Sun CJ, Yeh CC. The thermotreatment of msw incinerator fly ash for use as an aggregate: a study of the characteristics of size fractioning. Resource Conserv Recycling 2002;35:177–90.

[11] Ramachandran VS. Waste and by-products as concrete aggregates. Canadian building digest CBD-215 National Research Council, Canada; 1981.

[12] Ramamurthy K, Harikrishnan KI. Influence of binders on properties of sintered fly ash aggregate. Cem Concr Compos 2006;28(1):33–8.

[13] Manikandan R, Ramamurthy K. Swelling characteristics of bentonite on pelletization and properties of fly ash aggregates. J Mater Civil Eng 2009;21(10):578–86.

[14] Yoo JG, Jo YM. Finding the optimum binder for fly ash pelletization. Fuel Process Technol 2003;81(3):173–86.

[15] Bijen JMJM. Manufacturing processes of artificial lightweight aggregates from fly ash. Int J Cem Compos Lightweight Concr 1986;8(3):191–9.

[16] Swamy RN, Lambert GH. The microstructure of Lytag aggregate. Int J Cem Compos Lightweight Concr 1981;3(4):273–82.

[17] Wasserman R, Bentur A. Effect of lightweight fly ash aggregate microstructure on the strength of concretes. Cem Concr Res 1997;27(4):525–37.

[18] Verma CL, Handa SK, Jain SK, Yadav RK. Techno-commercial perspective study for sintered fly ash light-weight aggregates in India. Construct Build Mater 1998;12(6–7):341–6.

[19] Al-Khaiat H, Haque MN. Effect of initial curing on early strength and physical properties of a lightweight concrete. Cem Concr Res 1998;28(6):859–66.

[20] Al-Khaiat H, Haque N. Strength and durability of lightweight and normal weight concrete. J Mater Civil Eng 1999;11(3):231–5.

[21] Yun B, Ratiyah I, Basheer PAM. Properties of lightweight concrete manufactured with fly ash, furnace bottom ash, and Lytag. In: International workshop on sustainable development and concrete technology. Beijing; 2004. p. 77–88.

[22] Kayali O, Haque MN, Zhu B. Some characteristics of high strength fiber reinforced lightweight aggregate concrete. Cem Concr Compos 2003;25(2):207–13.

[23] Kayali O, Haque MN, Zhu B. Drying shrinkage of fibre-reinforced lightweight aggregate concrete containing fly ash. Cem Concr Res 1999;29(11):1835–40.

[24] Zhang MH, Gjorv OE. Mechanical properties of high strength lightweight concrete. ACI Mater J 1991;88(3):240–7.

[25] Harikrishnan KI, Ramamurthy K. Influence of pelletization process on the properties of fly ash aggregates. Waste Manage 2006;26(8):846–52.

[26] Döven AG. Lightweight fly ash aggregate production using cold bonding agglomeration process. PhD thesis. Istanbul, Boğaziçi University; 1998.

[27] Manikandan R, Ramamurthy K. Influence of fineness of fly ash on the aggregate pelletization process. Cem Concr Compos 2007;29(6):456–64.

[28] Chiou I, Wang K, Chen C, Lin Y. Lightweight aggregate made from sewage sludge and incinerated ash. Waste Manage 2006;26(12):1453–61.

[29] Li Y, Wu D, Zhang J, Chang L, Wu D, Fang Z, et al. Measurement and statistics of single pellet mechanical strength of differently shaped catalysts. Powder Technol 2000;113(1–2):176–84.

[30] Yashima S, Kanda Y, Sano S. Relationship between particle size and fracture energy or impact velocity required to fracture as estimated from single particle crushing. Powder Technol 1987;51(3):277–82.

[31] Diamond S. Mercury porosimetry: an inappropriate method for the measurement of pore size distributions in cement-based materials. Cem Concr Res 2000;30(10):1517–25.

[32] Moon HY, Kim HS, Choi DS. Relationship between average pore diameter and chloride diffusivity in various concretes. Construct Build Mater 2006;20(9):725–32.

[33] Yang CC. On the relationship between pore structure and chloride diffusivity from accelerated chloride migration test in cement-based materials. Cem Concr Res 2006;36(7):1304–11.

[34] Xue Y, Wu S, Hou H, Zha J. Experimental investigation of basic oxygen furnace slag used as aggregate in asphalt mixture. J Hazard Mater 2006;138(2):261–8.

[35] Al-Bahar S, Bogahawatta VTL. Development of lightweight aggregates in Kuwait. Arab J Sci Eng 2006;31(1C):231–9.

[36] Tsai C, Wang K, Chiou I. Effect of SiO_2–Al_2O_3–flux ratio change on the bloating characteristics of lightweight aggregate material produced from recycled sewage sludge. J Hazard Mater 2006;134(1–3):87–93.

[37] Mun KJ. Development and tests of lightweight aggregate using sewage sludge for nonstructural concrete. Construct Build Mater 2007;21(7):1583–8.

[38] Duvarcı OC, Akdeniz Y, Ozmihci F, Ulku S, Balkose D, Ciftcioglu M. Thermal behaviour of a zeolitic tuff. Ceram Int 2007;33(5):795–801.

[39] Aineto M, Acosta A, Rincon JM, Romero M. Thermal expansion of slag and fly ash from coal gasification in IGCC power plant. Fuel 2006;85:2352–8.

[40] Adell V, Cheeseman CR, Ferraris M, Salvo M, Smeacetto F, Boccaccini AR. Characterising the sintering behaviour of pulverised fuel ash using heating stage microscopy. Mater Character 2007;58(10):980–8.

[41] Adell V, Cheeseman CR, Doel A, Beattie A, Boccaccini AR. Comparison of rapid and slow sintered pulverised fuel ash. Fuel 2008;87:187–95.

[42] Biernacki JJ, Vazrala AK, Leimer HW. Sintering of a Class F fly ash. Fuel 2008;87(6):782–92.

[43] Styszko-Grochowiak K, Golas J, Jankowski H, Kozinski S. Characterization of the coal fly ash for the purpose of improvement of industrial on-line measurement of unburned carbon content. Fuel 2004;83:1847–53.

[44] Huang S, Chang F, Lo S, Lee M, Wang C, Lin J. Production of lightweight aggregates from mining residues, heavy metal sludge, and incinerator fly ash. J Hazard Mater 2007;144(1–2):52–8.

[45] Ilica M, Cheeseman C, Sollars C, Knight J. Mineralogy and microstructure of sintered lignite coal fly ash. Fuel 2003;82:331–6.

[46] Riley CM. Relation of chemical properties to the bloating of clays. J Am Ceram Soc 1951;34(4):121–8.