



Utilization of lignite power generation residues for the production of lightweight aggregates

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ABSTRACT

A novel process is proposed for the utilization of lignite combustion solid residues in the production of inflammable lightweight aggregates (LWA). The process consists of two stages, pelletization and sintering, and carbon contained in BA was used as the process fuel. The main residues bottom ash (BA) and fly ash (FA) from Megalopolis power plant were characterized, mixed in different proportions and treated through pelletization and sintering process. Sintering benefits from combustion of BA carbon content and the product is a hardened porous cake. The energy required for achievement of high temperatures, in the range of 1250 °C, was offered by carbon combustion and CO₂ evolution is responsible for porous structure formation. Selected physical properties of sintered material relevant to use as lightweight aggregates were determined, including bulk density, porosity and water absorption. Bulk density varies from 0.83 to 0.91 g/cm³, porosity varies from 60% to 64% and water absorption varies from 66% to 80%. LWA formed is used for the production of lightweight aggregate concrete (LWAC). Thermal conductivity coefficient varies from 0.25 to 0.37 W/mK (lower than maximum limit 0.43 W/mK) and compressive strength varies from 19 to 23 MPa (higher than minimum limit 17 MPa). The results indicate that sintering of lignite combustion residues is an efficient method of utilization of carbon containing BA and production of LWA for structural and insulating purposes. Carbon content of BA is a key factor in LWA production. Finally, this research work comprises the first proposed application for utilization of BA in Greece.

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1. Introduction

Nowadays, a large part of Greece's energy demand is covered by lignite combustion in West Macedonia and Peloponnesus. This energy is produced exclusively in plants of Public Power Corporation (PPC) and results in the production of over 14 Mt/year solid byproducts. Flying ash (FA) holds the 75% of the total production while bottom ash (BA) the rest. These byproducts are produced (4 Mt/year) at PPC plants in Megalopolis (Peloponnesus). Since 1982, the 12%, approximately [1] of the production of Megalopolis FA is utilized in the production of Portland cement the so-called Greek type FA-cement. In parallel FA from Ptolemais plants (Macedonia) has already been used in large construction projects such as the Platanovrisis Dam at Nestos. Lignite BA has not found any application yet, mainly due to high percentage of carbon contained. Therefore, it is of great importance to develop processes for utilization of the above byproducts especially directed to mas-

sive applications, such as the production of lightweight aggregates (LWA).

The annual production of LWA in Greece is 800,000 t in the form of natural pumice which is quarried from Giali island (Dodekanisa). To the present, the production covers the domestic market demands while it allows exports. In future, quarrying works should be reduced and eventually stopped, due to archaeological findings in the area and destruction of natural environment of the island.

At the same time, LWA demands increase, as a result of their expanding applications as structural—insulating materials, as ground/garden sub layers in land drainage applications, in floor and roof thermal and sound insulations, etc. This demand will be intensified in the future due to energy savings and inflammability advantage that the application of these materials offers. Significant sector of research activity is focused in the production of LWA using industrial wastes and byproducts as raw materials mainly by sintering processes.

In the production process at sintering temperatures, usually over 900 °C [2,3], LWA are formed due to a bloating effect that takes place [4]. This is an expansion phenomenon in which development of a glassy phase and evolutions of gases exist simultaneously, and

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results to the production of material with high porosity and low density.

Industrial solid wastes from power and municipal waste incinerator plants have already been used for production of LWA's. Bhatti and Reid [5,6] used incinerator sewage sludge ash as raw material for production of lightweight aggregates. The material is pelletized and sintered at 1060–1100 °C. The bulk density of sintered product ranges from 0.48 to 0.67 kg/m³. The compressive strength of lightweight aggregate concrete (LWAC), produced with the addition of this LWA, has maximum value 12 MPa for concrete density 1600 kg/m³. Biernacki et al. [7] performed sintering tests with coal fly ash at 1050–1200 °C with different processing times and determined the best product quality according to physical properties relevant to use as LWA, including density, strength, shrinkage and water absorption. Cheeseman et al. [8] used bottom ash from a municipal waste incinerator plant as the feed material in sintering process at temperatures 1000–1050 °C, and produced LWA with similar properties to the commercially available Lytag. Huang et al. [9] used mining residues, incinerator fly ash and heavy metal sludge ash as feed materials to the sintering process for LWA production. The results set the optimum (lowest specific gravity) condition for LWA fabrication at 1150 °C for 15 min with raw pellets fed at 750 °C. Tsai et al. [10] performed sintering tests using sewage sludge ash by altering percentages of SiO₂, Al₂O₃ and fly ash. The variation of each of these compounds in the mixture influence the physical properties (bulk density, strength) of the sintered final product.

A novel process is proposed for the utilization of Megalopolis lignite combustion byproducts in the production of LWA by a two-step process, pelletizing and sintering. LWAC was produced with the addition of the above material. Its properties, such as compressive strength and thermal conductivity, were measured.

For the sintering process carbon content of BA is utilized as a fuel. The fuel quantity in the sintered mixtures is controlled by the addition of FA. Small amount (3 wt%) of metallurgical lime is used to facilitate the formation of the pellets. In order to examine the potential of BA carbon content as a fuel two different type of BA are used, rich and poor in C, respectively. In the latter case, anthracite is added. For pelletization process the water content of BA, usually in the range of 30–40%, is used, with minor addition. This process is the first one proposed for the utilization of BA in Greece.

2. Materials and methods

2.1. Characterization of raw materials and sintering mixtures

The raw materials used for the experiments are two different types of bottom ashes, BA and BA_L (low carbon BA) rich and poor in carbon, respectively, FA as well as metallurgical lime and anthracite.

Chemical analysis after drying of raw materials is presented in Table 1. All ashes BA, BA_L and FA are classified as type F ones due to high total percentage of SiO₂, Al₂O₃ and Fe₂O₃ (higher than 70%) according to ASTM C 618 specifications [11]. BA and BA_L differentiate mainly to their carbon content 10.1 and 6.5 wt%, respectively, while FA remains 1.31 wt% C. Metallurgical lime was used as a binder to assist the formation of green pellets. The mixtures prepared and the proportions of the raw materials used are presented in Table 2.

In order to examine the amount of carbon required for sintering process, eight mixtures containing from 3.5 to 9.0 wt% are prepared by mixing BA and FA (mixtures M11 to M18). The efficiency of BA carbon content as a fuel was examined by replacing equal quantities of rich in carbon BA with poor BA_L. Anthracite was added in predetermined quantities so that the obtained mixtures M'13, M'15

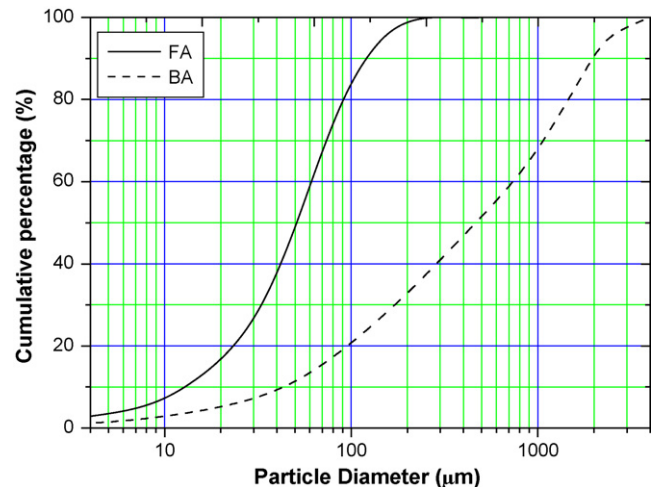


Fig. 1. Particle analysis of FA and BA.

and M'18 to have the same total carbon content as M13, M15 and M18, respectively.

In Fig. 1 the cumulative curves for the particle size distribution of FA and BA are depicted. FA is characterized as a fine-grained material particle size under 400 µm. The 70 wt% of the FA has particle size between 200 and 10 µm. BA is considerably coarser with maximum diameter of agglomerates 4000 µm. In this case the 10 wt% approximately is over 2000 µm, while under 10 µm is the 3 wt%.

Typical X-ray diffraction spectra for BA and FA are presented in Fig. 2. The reflection peaks of quartz (SiO₂), anorthite (CaAl₂Si₂O₈), gelhenite (Ca₂Al₂SiO₇), anhydrite (CaSO₄), hematite (Fe₂O₃) and aluminum oxide (Al₂O₃) are identified. The main difference between FA and BA is that in the later case the peaks of calcite (CaCO₃) are identified.

Fig. 3 shows the loss of ignition of BA and FA resulted from thermo-gravimetric analysis. Carbon combustion and decomposition of sulphur compounds (CaSO₄) is occurring from 300 to 700 °C. This phenomenon is confirmed by Fig. 4 which shows the emissions of CO and SO₂ during ignition up to 1000 °C. Decomposition of CaCO₃ is possibly achieved between 600 and 700 °C [12–14]. Furthermore, increase of weight above 800 °C is possibly explained by oxidation of FeO, which is formed in lower temperatures (<700 °C)

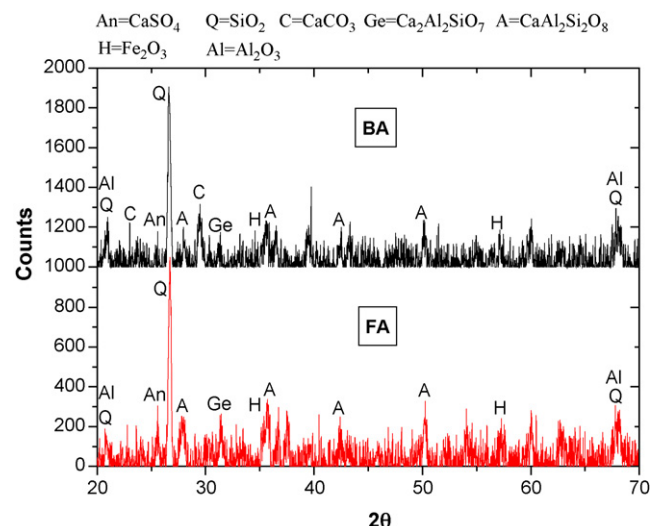


Fig. 2. XRD spectra of raw materials (FA and BA).

Table 1
Chemical composition of dry FA, BA and BA_L (wt%)

Oxide	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	C	Total
FA	49.4	22.65	7.42	8.85	1.62	0.85	1.39	2.55	1.31	96.04
BA	44.6	19.93	6.28	7.51	1.43	0.75	1.22	2.24	10.1	94.10
BA _L	46.2	20.82	6.54	8.34	1.49	0.81	1.27	2.41	6.52	94.40

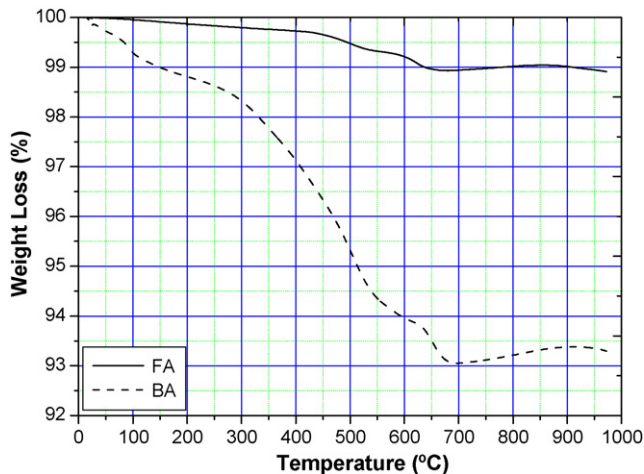


Fig. 3. Thermogravimetric analysis of FA and BA.

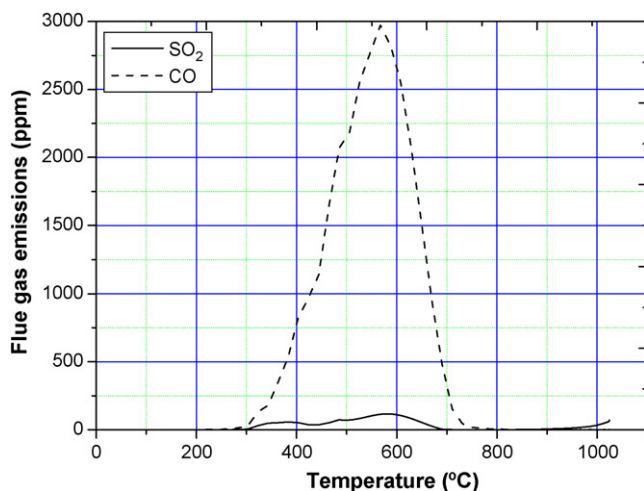


Fig. 4. Flue gas emissions of BA.

due to reduction environment [15]. Formation of FeO promotes formation of eutectic compounds up to 1000 °C. A further weight loss between 900 and 1000 °C is caused by SO₂ evolution (Fig. 4), probably as a result of decomposition of alkali metal sulphates [12,16,17].

Table 2
Sintering mixtures composition

Mixture symbol	FA (wt%)	BA (wt%)	BA _L (wt%)	Anthracite (wt%)	M. lime (wt%)	Total contained 'C' in mixture (wt%)
M11	62	35	–	–	3	3.5
M12	42	55	–	–	3	5.5
M13	32	65	–	–	3	6.5
M14	27	70	–	–	3	7.0
M15	22	75	–	–	3	7.5
M16	17	80	–	–	3	8.0
M17	12	85	–	–	3	8.5
M18	7	90	–	–	3	9.0
M'13	29.7	–	65	2.3	3	6.5
M'15	19.3	–	75	2.7	3	7.5
M'18	3.8	–	90	3.2	3	9.0

2.2. Experimental

The developed process for the production of LWA from lignite ashes consists of two stages, pelletization and sintering, respectively.

Prior pelletization, BA, BA_L and FA dusts are thorough mixed in pots with addition of 3 wt% fine-grained metallurgical lime as binder and anthracite as extra fuel in the case of low C mixtures M'13, M'15 and M'18. Subsequently, 3–10 mm in diameter pellets are formed by means of a pelletizing disk rotating at 12–15 rpm at a slope of 45° [18,19]. Minor water addition is required for the formation of the pellets due to the relatively high water content of BA, which is usually in the range of 30–40%.

For the sintering process, the pellets are loaded in a sinterpot of 0.04 m² surface so to form a 0.20–0.25 m height air permeable bed. The process is started with the ignition of a charcoal layer placed at the surface of the bed. Air is drawn downwards through the grate, by means of an exhaust blower connected from underneath to the grate. In this way, suction pressure is created through the bed. The suction pressure was adjusted to 10–15 bar. The narrow combustion zone developed initially at the top layer travels through the bed raising the temperature of the bed layer by layer, to the sintering level. In the combustion zone, bonding takes place between the grains and a strong and porous aggregate is formed. The process is over when the combustion zone reaches the lowest layer of the bed [20].

The temperature across the bed is recorded by three type K thermocouples placed vertically to the height of the bed. The two thermocouples T₁ and T₂, are placed 15 and 100 mm, approximately, from the upper surface of the bed, respectively. The third one T₃, is recording the temperature at the lower level of the bed, just above the grate, approximately 200 mm from the surface. The experimental installation with the pelletizing disc and sinter pot devices is schematically presented in Fig. 5. In Fig. 6, view of part of the opened pot with the sinter cake in it is depicted.

3. Results and discussion

3.1. Sintering products

All mixtures presented in Table 2 are sintered in the sinter pot. It is observed that sintering takes place for mixtures with carbon content over 6.5 wt%. Sintering is not possible for mixture

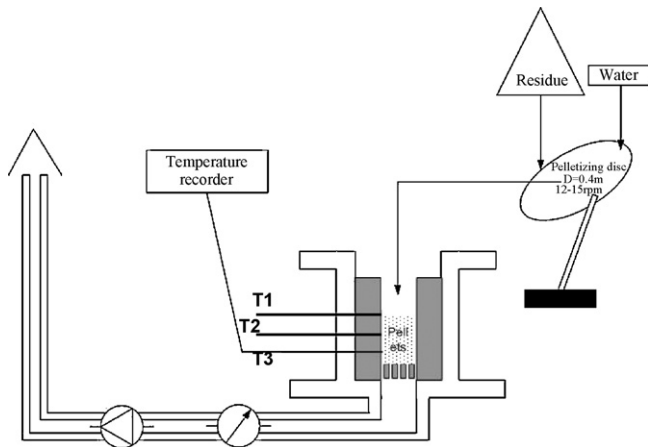


Fig. 5. Schematic diagram of the experimental installation.

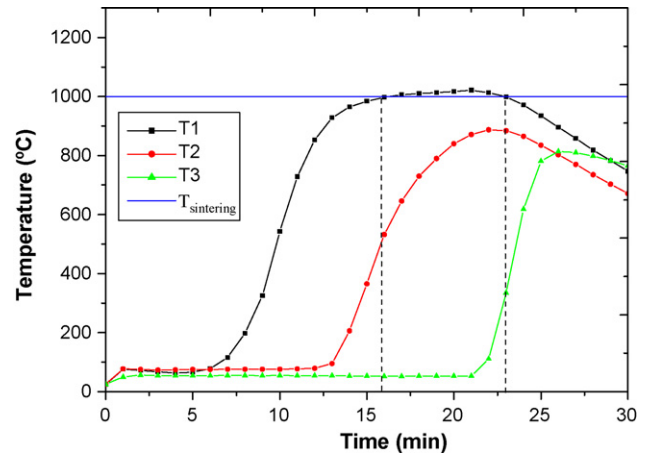


Fig. 8. Temperature to time curve for mixture M11.



Fig. 6. View of part of the opened sinterpot with the formed sinter cake inside.

M11 with 3.5 wt% C while mixture M12 with 5.5 wt% C is partially sintering.

In Fig. 7 the temperature distribution during sintering of mixture M13 is observed. Similar behavior is observed in all cases where sintering takes place. In all these cases, temperature in the bed exceeds 1000 °C. For mixture M11 temperature is lower than 1000 °C as Fig. 8 shows. Therefore, the above results indicate that

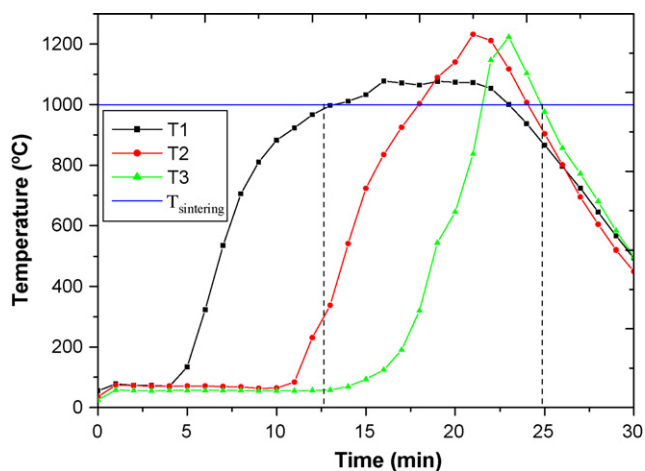


Fig. 7. Temperature to time curve for mixture M13.

sintering requires temperatures over 1000 °C and carbon content over 6.5 wt% C.

Chemical analysis of the produced sinters is presented in Table 3. The carbon content of sintered products is lower than 1 wt%. The low carbon percentage indicates that almost total quantity of the solid fuel is consumed during sintering. In large-scale operation, 100% consumption of the solid fuel is anticipated.

All sintered products present similar crystalline structure. The results from the XRD analysis for the sintered products are presented in Fig. 9. The intensity of quartz peaks (Q) is decreased, as the peaks of anorthite (A) are increased, in all mixtures tested. Furthermore, cristobalite (CB) and wollastonite (CS) are identified and reduction of hematite (H) to magnetite (M) is observed. Formation of new compounds such as A, CS and CB has also been reported by other researchers [21–23] as the result of FA sintering, with similar chemical composition, in high temperatures (~1250 °C).

Microstructure analyses are performed using a scanning electron microscope (SEM) and Fig. 10 shows cross-section of two sintered pellets. Open and closed pores of different size and shape are observed, as a result of CO₂ evolution, mainly carbon oxides from carbon burning and the simultaneous formation of glassy phase. Other phenomena that contribute to gases evolution are Fe₂O₃ reduction, decomposition of limestone and SO₂ evolution [4].

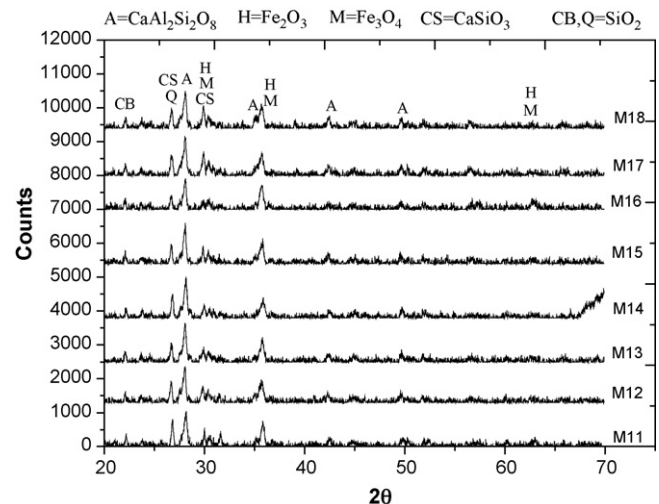


Fig. 9. Crystalline peaks of sintered products at 1200–1200 °C.

Table 3
Chemical composition of sintered products (wt%)

Oxides	M11	M12	M13	M14	M15	M16	M17	M18	M'13	M'15	M'18
SiO ₂	49.81	51.28	49.27	50.10	49.61	51.45	48.17	49.30	48.23	48.43	49.40
Al ₂ O ₃	23.86	24.18	25.55	25.60	24.52	23.20	26.01	23.06	24.61	24.02	23.72
Fe ₂ O ₃	7.09	7.01	7.80	6.30	6.69	8.41	8.13	6.63	7.30	6.42	6.57
CaO	10.92	12.04	12.68	12.80	10.68	12.68	9.54	10.45	11.84	10.92	10.33
MgO	2.02	1.75	1.95	2.01	2.01	1.81	2.23	1.97	1.88	1.82	1.90
Na ₂ O	1.37	1.26	1.46	1.80	1.67	1.36	2.00	1.73	1.26	1.52	1.64
K ₂ O	1.54	0.78	0.98	0.80	1.75	0.45	0.58	1.51	1.08	1.42	1.39
C	0.37	0.60	0.00	0.00	0.42	0.32	1.71	0.87	0.46	0.63	0.91
Total	96.98	98.90	99.70	99.41	97.35	99.70	98.30	97.52	96.66	95.18	95.86

Apparent porosity, particle density and water absorption are measured for all mixtures aggregates. For this purpose, sintered cake is mechanically crushed into aggregates with a size from 8 to 16 mm. The measurements are performed according to ASTM C373–88 specifications [24] and the results are presented in Fig. 11a–c. Apparent porosity values vary between 60% and 64% (Fig. 9a) and the maximum one is shown for M13 (65 wt% BA, 6.5 wt% carbon). Bulk density of all mixtures is below 1 g/cm³ (Fig. 9b). The minimum density value (0.83 g/cm³) is achieved for aggregates with 65 wt% BA. The maximum water absorbed (80%) is achieved also for mixture with 65 wt% BA.

The above results indicate that sintering of mixture M13 results to lightweight materials with the best performance among the tested ones as far as it concerns the most important physical properties for LWA. Carbon content of 6.5 wt% seems to be enough for successful sintering and porous structure formation, without excess densification of the product.

Fig. 12a–c presents comparison of measured values of mixtures M'13, M'15 and M'18 to M13, M15 and M18. Products of mixtures without anthracite show higher porosity, water absorption and lower bulk density of particles than products of mixtures with anthracite. Products without anthracite show porosity values from 60% to 64%, bulk density values from 0.83 to 0.91 g/cm³ and water absorption values 66–80%. Products with anthracite show porosity values from 48% to 54%, density values from 1.01 to 1.08 g/cm³ and water absorption values from 45% to 54%. Sintered products without anthracite show improved physical properties, relevant to use as LWA, compared to products with anthracite. Therefore, BA carbon combustion is a key factor for LWA production. This is attributed to the fineness and high degree of uniformity of the carbon contained in BA. Combustion of fine and wide dispersed

carbon grains leads to the formation of a homogeneous porous structure consisting of a large number of small size pores. On the other side, anthracite is difficult to be evenly dispersed in the mixture and thus, carbon distribution does not attain the same degree of uniformity in the mixture. As a result, bigger and open pores are formed with non-uniform distribution and heavier lightweight aggregates are produced.

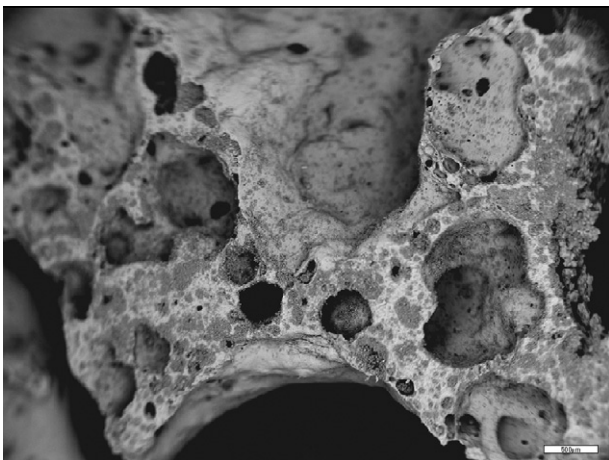
3.2. Physical and mechanical properties of LWAC

Of great importance for the practical application of the so produced artificial LWA are the physical and mechanical properties of the LWAC. Therefore, LWA produced from sintering are used for the production of LWAC. Specimens of different mixture and size are prepared for measurements of their coefficient of thermal conductivity (ctc) and compressive strength. All specimens contain two different fractions of aggregates: 0–5 and 8–16 mm. The 0–5 mm fraction is pumice for all specimens. The 8–16 mm fraction is artificial lightweight aggregate produced from sintering. Specimen with pumice of 8–16 mm fraction is produced, as well, for direct comparison with the artificial lightweight aggregate specimen. Pumice of 8–16 mm fraction has a bulk density of 550 kg/m³ according to ASTM C29M-7 specifications [25] and particle density of 0.96 g/cm³ [24].

The specimen dimensions for ctc measurements are 200 mm × 200 mm × 45 mm. The materials used for these specimens are: LWA 881 kg/m³, water 198 kg/m³ and cement 224 kg/m³, with w/c=0.8. Fifty percent of each aggregate fraction is used. The specimen dimensions for compressive strength tests are 150 mm × 150 mm × 150 mm. The materials used are: LWA 756 kg/m³, water 195 kg/m³ and cement 320 kg/m³, with w/c=0.61. Aggregates size distribution is 70% of 0–5 mm fraction and 30% of 8–16 mm fraction.

3.2.1. Thermal conductivity

LWAC specimens are prepared for sintered mixture M12–M18 and the ctc is measured in three different temperatures: –15, 20 and 70 °C, respectively. The measurements are carried out by using the method of hot plate and the equipment incorporated is a FOX

**Fig. 10.** Cross-section of sintered pellets.**Table 4**
Twenty-eight days oven dry density of LWAC specimens

Specimen of mixture	28 days dry density (kg/m ³)
M12	1362
M13	1356
M14	1371
M15	1368
M16	1348
M17	1354
M18	1342
Pumice	1344

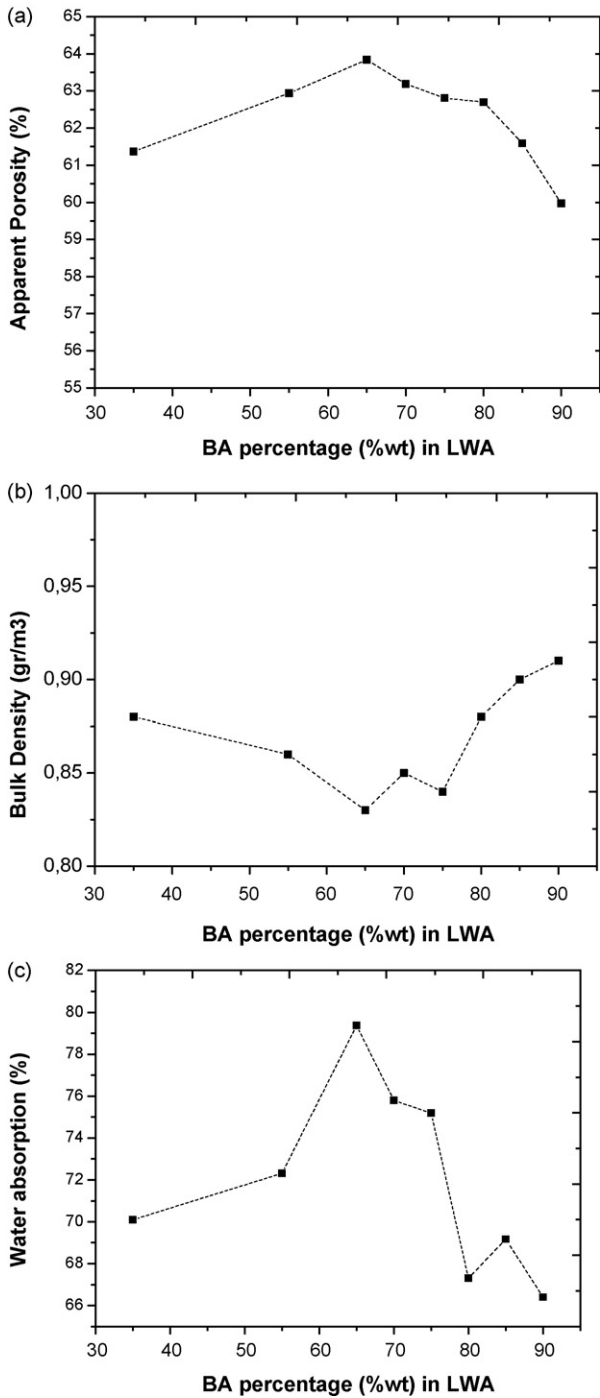


Fig. 11. (a) Apparent porosity, (b) bulk density, (c) water absorption dependence of BA carbon content of sintering mixtures M11–M18.

200 of Lasercomp. This device is operating according ASTM C518 and ISO 8301 specifications. The specimens are cured for the first 7 days at 20 °C and 95% humidity and for the next 21 days at room temperature and humidity. Oven dry density of 28 days is measured for all specimens. The values are presented in Table 4.

The measured values of *ctc* of all specimens at all three temperatures are presented in Fig. 13. All values of artificial LWAC specimens at –15 °C (0.25–0.31 W/mK) approximate that of pumice LWAC, while mean value coincides with pumice LWAC (0.28 W/mK). The values of *ctc* at 20 °C are between 0.27 and 0.36 W/mK. The maximum deviation of artificial LWAC *ctc* from the pumice spec-

imen is 0.03 W/mK and their mean value almost coincides with pumice LWAC. The values of *ctc* at 70 °C are between 0.28 and 0.37 W/mK and deviations from pumice specimen are higher (up to 0.06 W/mK). Mean value of artificial LWAC coincides with pumice LWAC value (0.34 W/mK). According to ASTM C 332-9 specifications [26] the limit for insulating lightweight concrete is 0.43 W/mK for maximum oven dry density 1440 kg/m³. As a result, all the specimens produced are clearly below (~20%) these limits, and the LWA used are suitable for production of insulating lightweight aggregate concrete.

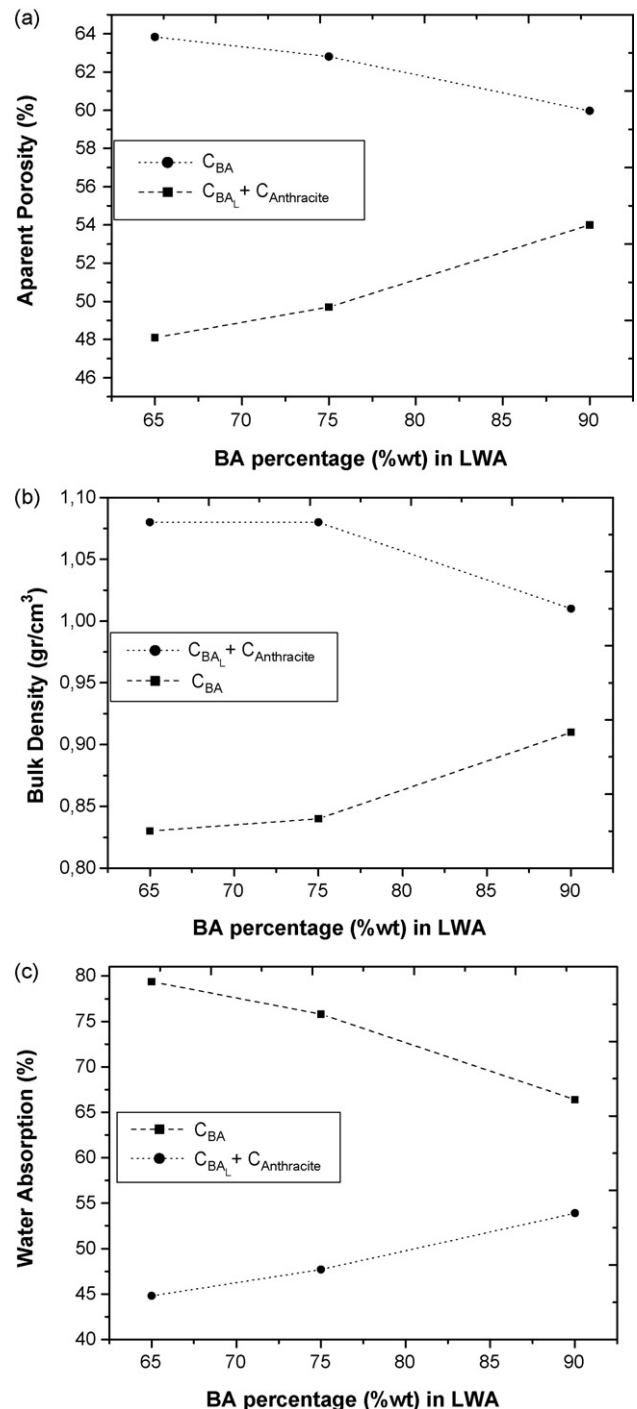


Fig. 12. (a) Apparent porosity, (b) bulk density, (c) water absorption dependence of BA carbon and anthracite of sintering mixtures M13, M15 and M18.

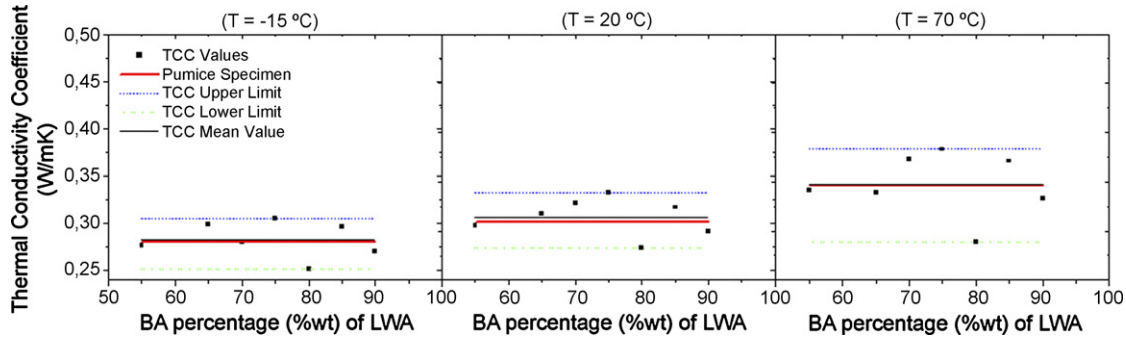


Fig. 13. Thermal conductivity coefficient at –15, 20 and 70 °C for LWAC of mixtures M12–M18 and pumice.

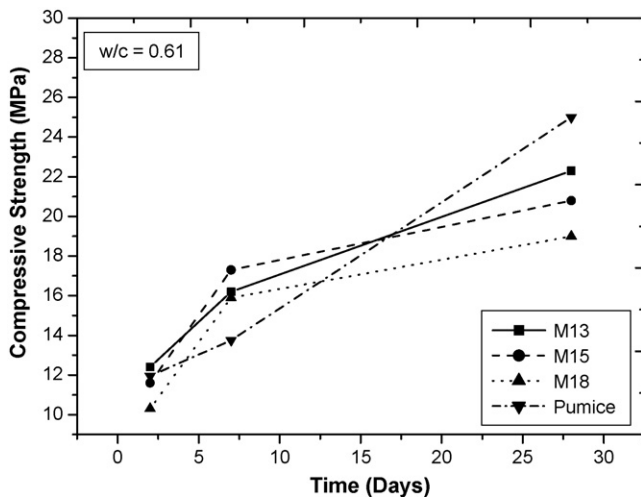


Fig. 14. Compressive strength of LWAC of mixtures M13, M15 and M18.

3.2.2. Mechanical strength

The results from compressive strength tests of LWAC specimens are presented in Fig. 14. Cubic specimens are prepared for mixtures M13, M15 and M18 and for pumice, as well. The specimens were cured at 20 °C and 95% humidity until the day of strength measurement (2, 7 and 28 days). The compressive strength measurement of cubic specimens is carried out by using a machine of 1600 kN capability.

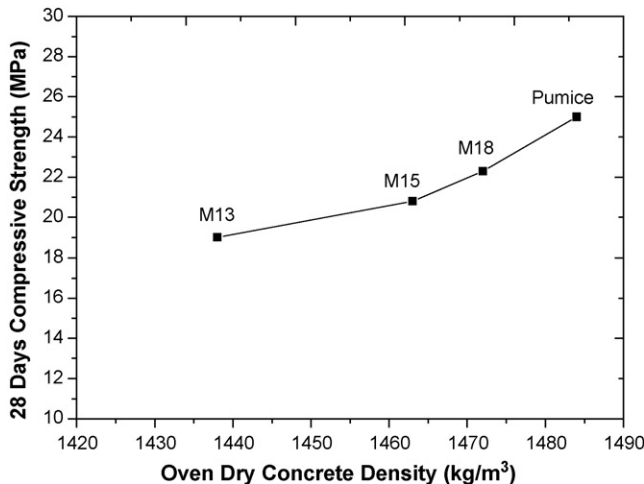


Fig. 15. Relationship between strength and LWAC density.

Pumice shows the highest strength value 25 MPa at 28 days, while it has the lowest value at 7 days, 13.8 MPa. The strength values of artificial LWAC at 28 days range from 19 to 23 MPa. In Fig. 15 is presented the relationship between 28 days strength and specimens dry density. Increase in density is directly related with increase in compressive strength. Furthermore, the lower limit for lightweight aggregate structural concrete at 28 days is 17 MPa (ASTM C 330-97) [27] with average air dry density 1600 kg/m³. The values of specimens' strength of all mixtures are above this limit. As a result, the LWA's produced from sintering of mixtures M13, M15 and M18 can be used for structural purposes.

4. Conclusions

In purpose of utilization of lignite combustion solid residues in LWA production, mixtures of BA and FA are sintered at 1200–1250 °C and the BA carbon content is used as the solid fuel of the sintering process. Sintering tests of different proportions of BA and FA proved that carbon content of 6.5 wt% in the sintering mixture elevate temperature above 1100 °C, necessary for successful sintering. During sintering, burning of BA fine-dispersed carbon and evolution of CO₂ inside the sintering pellets leads to the formation of LWA with high porosity from 60% to 64%, bulk density from 0.83 to 0.91 g/cm³ and high water absorption from 66% to 80%. Alteration of carbon content results in fluctuation of final product physical properties, including density, porosity and water absorption. Partial substitution of BA carbon by fine anthracite, in the mixture, results in deterioration of produced LWA physical properties. Thus, carbon content of BA is a key factor in LWA production, through sintering process, by means of energy provide and porous structure formation. In addition, an important advantage of this method is that the raw materials can be used without any pre-treatment (milling, drying).

The LWA formed are used for production of LWAC. Thermal conductivity and compressive strength measurements of LWAC indicate that these new materials can be used for insulating and structural purposes. Thermal conductivity coefficient of all LWAC specimens is range from 0.25 to 0.37 W/mK (lower than the maximum value 0.43 W/mK). Compressive strength values of all LWAC specimens produced, range from 19 to 23 MPa (higher than the minimum strength value 17 MPa).

This research work opens a new way for lignite BA utilization. Similar methods have not introduced until now in Greece. Furthermore, it was shown that the carbon content of lignite BA is a serious advantage in production of LWA.

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