Assessment of the mineral phase and properties of clay-Ca bentonite lightweight aggregates

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Abstract

This study deals with improving the properties of lightweight aggregate by the addition of Ca-bentonite mineral to expandable clays. The addition of bentonite can result in advanced lightweight aggregate materials which meet international standards. Besides bentonite material, twelve different clays (from Egyptian and Hungarian mines) were studied by X-ray fluorescence (XRF), X-ray powder diffraction (XRD, Rietveld method), and a heating microscope (HM). Aggregates were produced and their physical and mechanical properties were measured. The structure and the chemical composition of the clay-Ca bentonite aggregates were studied by scanning electron microscope (SEM) and with energy dispersive spectroscopy (EDAX). The results showed that the addition of 10 wt% bentonite can reduce the bulk density by up to 20%, can increase the compressive strength by up to 180%, and can increase the height of expansion by up to 68% in some samples. It was found that the high kaolinite content of the clay samples was not beneficial for expanding and decreasing the bulk density of aggregates. According to the Rietveld method from XRD and a statistical correlation model shown, the amorphous contents have a reciprocal relationship with the bulk density and a direct relationship with the height expansion. Bentonite, as a natural additive, to some clays, was suitable for producing lightweight aggregates with far better properties, as well as a more economical means of production.

Keywords Bentonite · Clay · Lightweight aggregate · Rietveld

Introduction

Due to their thermal performance, sound insulation, and good fire resistance, lightweight aggregates (LWA) are becoming the focus of interest all over the world. They are characterized as building materials that have a bulk density of less than 1.2 g/cm³, particle density in the range of 0.8–2 g/cm³, and compressive strength higher than 1 MPa (Choudhry and Hadley 1992; Liao and Huang 2011). Lightweight aggregates are made from materials such as clay, shale, slag, natural pumice, tuff, and perlite. To prepare LWA

from expandable clay, the aggregates must be expanded to approximately twice the volume of the original raw material.

During the industrial process, clay is crushed, screened to size, and finally fired in a rotary kiln. Firing temperatures are around 1300 °C (Rashad 2018; Alexander et al. 1999). A clinker cooler is used to decrease the temperature of the aggregates which are finally crushed and screened if necessary (Ozguven and Gunduz 2012; Chandra and Berntsson 2002).

The mechanism of expansion in LWAs, which takes place in several steps, was described by Riley (1951). The first step is the evolution of gasses at a certain temperature where the material is in its visco-plastic state. The sources of gas generation in this process can be the reduction of ferric oxide, the combustion of organic matters or the thermal decomposition of carbonates (Rudnai 1963; Chopra et al. 1964; Lee 2016). In the second step, a partially molten state of the clay prevents the escape of gases. Finally, after cooling, the molten phases remain as glassy phases containing different crystalline compounds depending on the composition of the raw material (Ozguven and Gunduz 2012). When

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the temperature is too high during sintering, excess gas can evolve through the surface of the aggregates, and this increases the overall porosity and creates continuous pores (Yang et al. 2015). In addition, further increases in temperature beyond the pyro-plastic range can lead to viscous flow, reducing porosity, and pore size (Dondi et al. 2016). Expanded aggregates produced by heat treatment have lower density than natural aggregates and result in lighter concrete products.

For improving the physical, mechanical, or expansion properties of lightweight aggregates, additive materials are required (Kocserha and Gömze 2010; Kocserha et al. 2018). Some of these additives, such as iron, can increase expansion, but bulk density and compressive strength are simultaneously reduced (Bernhardt et al. 2014).

With the addition of CaF_2 , growth in the glassy phase was achievable, but it decreased the sintering and melting temperature by 125–200 °C lower than the temperature used in the production of LWAs (Lee and Liu 2009; Abdelfattah et al. 2019).

In a previous experiment, carbon fibres were added to the expandable clay. This additive lowered the particle density, and resulted in higher overall porosity and compressive strength (Moreno-Maroto et al. 2017). Additives and their effects mentioned in related studies are summarized in Table 1.

In our study, bentonite as a natural additive was used to improve the properties of aggregates. Bentonites are clay soils formed by erosional action on volcanic ash, and consisting of montmorillonite, illite, and a variable composition of other minerals, such as quartz, sodium feldspar, and calcium feldspar according to the Rietveld method (Abu-Jdayil

2011). Structurally, bentonite is classified as a 2:1 layered aluminosilicate and ideally consists of an alumina octahedral sheet between two silica tetrahedral sheets (Ahmad et al. 2011; Paluszkievicz et al. 2008).

Bentonite is used in several branches of industry. It is used as oil drilling mud, a suspending agent in oil drilling fluids, or as a binder in the production of pellets (Grim 1968). It plays a role in the construction of municipal waste or spent nuclear fuel storage barriers (Pusch 1992; Gens et al. 1998; Yong 1999; Cui et al. 2002; Chegbeleh et al. 2008; Murray 2006; Pusch and Weston 2012). In calcined form, it affects the fresh and strength properties of mortar and concrete (García-García et al. 2006; Ibrahim et al. 2019), or in other studies, it was used as a substitute for cement (Taylor-Lange et al. 2015; Rajczyk and Langier 2012). In every case, it produced appropriate results.

In a preliminary study (Abdelfattah et al. 2020), three expandable Hungarian clays were mixed with 5, 10, and 20 wt% Ca-bentonite and the prepared aggregates were heat treated. The results showed that the addition of 10 wt% Ca-bentonite has a beneficial effect on most properties such as bulk density, compressive strength, and the height expansion of the aggregates.

The data in Table 1 show that researchers modified the properties of aggregates with artificial additives. In contrast, a natural additive, bentonite, was used in our experiment. The goals of this study were to produce LWAs from twelve expandable clays originating from different places and to observe and describe the effect of bentonite as a natural material addition on the properties of produced aggregates.

Table 1 The influence of the additive materials on the properties of LWAs from literature reviews

Author(s)	Additive types	Amount, %	Effect
Bernhardt et al. (2014)	Na_2CO_3	2	High degree of open porosity with poor expansion
	SiO_2	5	Increase in porosity
	Fe_2O_3 and Fe	5	Enhanced porosity and a high degree of expansion
Lee and Liu (2009)	CaF_2	10–20	Great influence on compressive strength
Wei et al. (2018)	CaF_2	10–20	Temperature decreases by 200 °C
Moreno-Maroto et al. (2017)	Carbon fibers	5–10	1-Drop in particle density
			2- Increase in total porosity
			3- Enhanced compressive strength
			4-Carbon fiber can achieve energy savings
Islam et al. (2016)	Soda-lime silica glass	20–50	1-Bulk density and water absorption decreased
			2- Improvement in crushing resistance of LWA
Lee (2016)	Amorphous carbon and kerosene	2–4–6	1- Decrease in density
			2- Lower expanding and sintering temperature
Liu et al. (2017)	Autoplastic	0.01	1-Increased porosity by 39.5%
			2- Bulk density decreased by 28.5%
Fakhfakh et al. (2007)	Quartz + automobile	15 + 1	1-Formation of more gas
			2-Drop in bloating temperature
			3-High water absorption capacity

Materials and methods

Clays and bentonite used for our experiments originated from Hungary and Egypt. Three types of clay samples (referred to as yellow (Y), grey (G), and blue (B)) were collected from the Mályi clay mine, in Hungary, and nine were taken from the Giza (marked with 1), Fayum (2), Sinai (5), Helwan (6), and Saf (8,9,10,11,12) deposits, in Egypt (Fig. 1). The bentonite originated from a deposit in Hungary (Istenmezeje). The clays were dried at 105 °C for 24 h in an electric drying chamber. They were ground to fit through a sieve with an aperture of 100 µm for further measurements (Liao and Huang 2011; Bernhardt et al. 2014).

For the chemical analysis of all of the major and minor oxides, and trace elements of the samples, a Rigaku Supermini 200 WDXRF instrument was used. The mineralogical composition of the clays and aggregates was determined by the X-ray diffraction method; measurement was carried out on a Rigaku Miniflex II instrument with Cu-K α radiation, $\lambda = 1.5418 \text{ \AA}$, in a range of $3^\circ \leq 2\theta \leq 90$. Phase identification and quantitative Rietveld refinements with the calculation of goodness of fit (GOF) were performed using the Profex 4.3 software. Differential thermal analysis (DTA) and thermogravimetric (TGA) measurements were also conducted

(MOM Derivatograph C). To observe the melting properties and dimensional changes of the clays, a heating microscope (Camera Electronica) was used up to 1400 °C with 10° C/min heating rate. Image analysis revealed the main characteristic temperatures of the sintering process.

After analysis of the raw materials, clays and 10 wt% Ca-bentonite and water were mixed, and green pellets having a diameter of 6–14 mm were prepared by the hand-rolling method and by wetting the clays (Bernhardt et al. 2014; Fakhfakh et al. 2007). They were dried at 105 °C in a drying chamber. This method was used for clays with and without additives. In order to determine the appropriate sintering temperature, raw aggregates without bentonite were sintered at five different temperatures (1150–1275 °C), in an air atmosphere. The sintering time was 10 min at maximum temperature. After the heating process, the sintered products were cooled naturally in the furnace.

For the Hungarian samples, the best properties were achieved at 1225 °C, and for the Egyptian samples at 1275 °C. These results are presented below.

A scanning electron microscope in high vacuum mode (Carl Zeiss EVO MA10) was used to identify the surface morphology of the different samples of the clay aggregates. Adhesive tape was used to hold the specimens in place on

Fig. 1 Location map for the clay samples **a** Hungarian sample. **b** Egyptian samples

the specimen holder and they were coated with gold according to ion beam sputtering.

The density and water absorption of the aggregates were measured according to the European standard (EN 13055-1 2002 and EN-1097-6:2013). The bulk density was calculated using Eq. (1)

$$\text{Bulk density} = W_D / (W_S - W_I) [\text{g/cm}^3] \quad (1)$$

Water absorption of the aggregates was determined according to Eq. (2)

$$\text{Water absorption} = 100 (W_D - W_S) / W_D [\%] \quad (2)$$

where W_D : the dry weight of the LWA, W_S : 24-h saturated surface-dry weight, W_I : immersed weight in water.

Similar to other studies (Hiramatsu and Oka 1966; Li et al. 2000; Liao and Huang 2011; Yang et al. 2015; Li et al. 2020), the compressive strength of the aggregates was measured on spherical aggregates individually. The aggregates were placed into a cylindrical mold, and an INSTRON universal tester with a cross-head speed of 0.1 mm/s was used to apply and register the fracture force. The results were calculated using the following formula (3)

$$\text{Uniaxial compressive strength (UCS)} = 2, 8F / \pi D^2 [\text{MPa}] \quad (3)$$

where F : load belongs to fracture, D : diameter of the aggregates.

Results and discussion

Characterization of raw materials

The chemical compositions of the clay samples are summarized in Table 2. Oxides were classified into three groups such as silica, alumina, and total flux, and they are plotted in the Riley diagram, Fig. 2a, (Riley 1951). The amount of SiO_2 in the clays ranged from 55.15 to 64.97%, of Al_2O_3 17.29–22.53%, the total flux varied between 15.96 and 21.24%. The iron contents of the samples are also indicated (6.3–14.21%). As Fig. 2a shows, each of the 12 clays was in the expanding range (red contour-encased area), which means that clay can be expanded when fired (Kavas et al. 2011; Anan and Abd El-Wahed 2017). Samples can obtain a suitable viscosity to trap a significant amount of the gaseous components, leading to the formation of the expanded structure of LWA (Hung and Hwang 2007).

After a 10% of bentonite addition to the clay sample, the amount of SiO_2 in clays ranged from 55.28 to 64.12%, Al_2O_3 17.53–21.93%, total flux 17.23–21.32%, and the iron content varied from 7.12 to 14.24%.

Table 2 Chemical composition of the clay samples

	Samples												
	Blue	Gray	Yellow	1 (Giza)	2 (Fayume)	5 (Sinai)	6 (Helwan)	8 (Saf)	9 (Saf)	10 (Saf)	11 (Saf)	12 (Saf)	Bentonite
SiO ₂ , %:	62.23	64.97	62.91	56.4	61.09	62.05	61.5	55.15	56.07	59.1	55.4	59.14	56.5
Al ₂ O ₃ , %:	17.73	17.36	17.94	20.75	19.33	19.54	19	22.18	22.03	18.48	21.47	17.29	19.7
Flux*, %:	19.31	17.43	18.3	21.24	17.53	16.75	16.9	20.04	19.6	19.18	20.35	20.22	20.2
Other oxides, %:	0.73	0.24	0.85	1.61	2.05	1.66	2.6	2.63	2.3	3.24	2.78	3.35	3.6
Fe ₂ O ₃ ** %,:	6.99	6.3	7.74	9.88	11.27	5.82	9.9	13.36	13.85	12.25	14.21	12.81	14.55

*Flux: $\text{CaO} + \text{Fe}_2\text{O}_3 + \text{Na}_2\text{O} + \text{K}_2\text{O} + \text{MgO}$, wt%

** Fe_2O_3 is also given separately and among the fluxes

Fig. 2 **a** Chemical composition of the technological sample as plotted on Riley's diagram (1951). **b** Chemical composition of the technological samples with 10% of bentonite as plotted on Riley's diagram (1951)

Fig. 3 DTA/TGA curves of the blue sample

Some of these values are plotted in the Riley diagram and presented in a magnified format in Fig. 2b. It can be seen that the points representing the different samples shifted toward the direction of fluxing oxides. Also, samples with bentonite contain more iron-oxide than the original raw materials.

A typical curve of DTA/TGA of the blue sample is shown in Fig. 3. Up to 550 °C, the DTA curve has the first wide exothermic peak which represents the burning out of the organic contents, and parallel to this, the decomposition of the structure of the clay minerals begins. At about 800 °C, the decompositions of carbonates ended. This is marked with point 1 on the TG curve. Above 1000 °C, a strong endothermic process begins which indicates that the melting of the sample begins. According to TG test, the total weight loss for the clay samples was between 8 and 19%.

The mineral phases of the collected clay samples were revealed by XRD. Results can be seen in Table 3. Analysis showed that the major mineral phases in all samples are quartz, illite, kaolinite, calcite, albite, and smectite. Other phases like muscovite, halite, orthoclase, goethite, clinocllore, and hematite were also found in the clays.

Characterization of clay samples with bentonite

Expansion extent

The melting behaviour of the clay was studied using a heating microscope. Photos were taken of all samples with and without bentonite additive at different temperatures (T). Image analysis revealed the height expansions (H) and the maximum height of expansion (MHE) of the clay samples (Li et al. 2020; Karaş et al. 2019). The MHE value indicates how much higher the height of the expanded sample is than that of the green sample. Besides the results, Fig. 4 shows the changes that occurred in the shape of sample 1 at different temperatures. For sample 1 from Egypt (Fig. 4a), the value of MHE was 129% at 1270 °C. After the addition of 10% of bentonite (Fig. 4b), the values of MHE increased to 135% at 1241 °C.

In the Riley diagram, a shift toward the direction of fluxing oxides due to the addition of bentonite was clearly observable, according to the results of heating microscope measurements. Based on the comparison of Fig. 4a and b, the temperature which belongs to MHE decreased by about 30 °C and the temperature at which a 10% increase in the height of the sample occurred was 75 °C lower with the addition of bentonite. Numerical values for all samples are summarized in the first columns in Table 4, while the results can be compared with the help of Fig. 5. The MHE value

Table 3 Mineral composition of the clay samples

Samples	Blue	Gray	Yellow	1 (Giza)	2 (Fayume)	5 (Sinai)	6 (Helwan)	8 (Saf)	9 (Saf)	10 (Saf)	11 (Saf)	12 (Saf)	Bentonite
Quartz	26.53	30.8	32.5	11.35	19.23	22.01	14.44	2.99	3.96	11.92	4.14	14.64	9.2
Illite	31.23	30.36	29.22	23.51	22.97	15.62	27.36	34.87	32.36	32.71	36.28	30.43	43.1
Kaolinite	2.57	3.2	1.68	27.23	18.35	22.87	21.74	33.46	33.3	31.04	32.09	15.81	
Albite	2.55	4.08	5.79	4.41	6.68	5.86	6.9	0.02	2.57	9.99	2.41	8.84	
Calcite	4.73	3.65	1.36	6.79	2.62	10.92	1.2	0.23	3.8	1.07	2.43	3.12	
Smectite	6.81	3.32	5.18	9.97	13.52	11.96	12.92	9.64	9.59		7.97	10.64	
Halite				0.97		0.8	0.26	0.57			0.53		
Orthoclase				3.72	3.5	2.6	1.66	2.56	2.56	1.48	1.59	3.45	
Goethite				2.05	5.28	0.5	0.78	0		1.27	0	1.06	
Dolomite	3.05	2.85	0.49										
Clinocllore	2.5	2.97	3.08										
Muscovite	10.21	12.52	7.71										
Rutile	0.82	0.24	1		0.85	0.84	2.76	6.65	5.85	3.79	6.31	7	
Hematite										0.73	0.25		
Vermiculite													17.2
Montmorillonite													24.2
Anorthite													6.3
Amorphous	9	6.01	11.99	10	7	6.02	9.98	9.01	6.01	6	6	5.01

Fig. 4 **a** The height expansion of clay sample (1) without additive at different temperatures. **b** The height expansion of clay sample (1) with 10% of bentonite at different temperatures

Table 4 Physical and mechanical properties of clay- Ca bentonite aggregates

Samples		MHE, %		BD, g/cm ³		UCS, MPa		WA, %
		Additive free	10% of bentonite	Additive free	10% of bentonite	Additive free	10% of bentonite	
Hungarian	B	138	164	0.57	0.55	1.54	3.27	0.7
	Y	122	135	1.2	0.95	4.82	7.15	0
	G	123	133	0.84	0.79	2.09	4.32	6.1
Egyptian	1	129	135	0.7	0.75	1.18	1.2	4.16
	2	122	129	0.95	0.94	1.1	2	6.4
	5	122	135	0.7	0.65	3.04	1.46	16.7
	6	122	124	0.99	0.95	5	4.2	7.14
	8	126	123	0.93	0.97	1.7	4.78	7.5
	9	112	132	0.9	0.99	4	1.2	30
	10	144	120	0.88	0.9	3.1	3.8	23.8
	11	123	133	0.89	0.98	5.8	4.78	4.3
	12	121	124	1.1	1.02	10.4	12.08	3.6

*MHE maximum height expansion, BD bulk density, UCS uniaxial compressive strength, WA water absorption, and B blue clay, Y yellow clay, G grey clay

of the Hungarian samples without bentonite was between 22 and 38% while for the Egyptian samples, it ranged from 12 up to 44%. After the addition of 10% of bentonite to the clays, the value of MHE was higher in most cases except for samples 8 and 10. For the Hungarian clays, MHE increased from 33 to 64% while the increase was between 20 and 35% for the Egyptian samples.

Bulk density and compressive strength

The bulk density for pelletized aggregates without any additive in this study was found to be less than 1.2 g/cm³. For the Hungarian samples, the range of density was between 0.57 and 1.2 g/cm³ at 1225 °C, while for the Egyptian samples

at 1275 °C, the lowest and the highest values were 0.66 and 1.11 g/cm³, respectively. Among samples without any additive, the blue clay sample had the lowest density. This was probably related to the greater amount of its low viscosity glassy phase during firing. The numerical values can be found in Table 4. The results of the compressive strength measurement were in the interval of 1.54–4.82 MPa for the Hungarian aggregates and 1.18–10.4 MPa for the Egyptian clay. All strength values are higher than 1 MPa, so these are strong enough to be used as LWAs (Liao and Huang (2011)).

Figure 6a, b show the change in bulk density and compressive strength before and after the addition of 10 wt% Ca-bentonite. The addition of bentonite had a great effect on the density and compressive strength of the aggregates.

Fig. 5 Bar chart for the height expansion before and after 10% of bentonite addition

The bulk density of the Hungarian samples changed to 0.55–0.95 g/cm³, which is a 4–26% decrease. With decreasing density, there was a significant improvement in compressive strength. The strength of the aggregates was 48–112% higher. This is a beneficial change for LWAs, as it can result in lighter building blocks and concrete elements with elevated strength.

The bulk density of the Egyptian clays with 10% bentonite changed in a different way, ranging from 0.65 to 1.02 g/cm³. The change was both decrease and increase depending on the sample. Compressive strength increased in five of the nine Egyptian clays tested, while it decreased by 18–70% in the four other cases. The change in density and compressive strength of the aggregates is related partly to the amount of kaolinite phase in the samples. Kaolinite was present in high percentages in samples 1, 8, 9, 10, and 11 (> 25 wt%). The compressive strength of sample 5 decreased due to its 10.92% calcite content. With increasing temperature, the decomposition of calcite increases CO₂ emission, causing more porosity and a decrease in density and compressive strength.

For the samples (8, 9, and 11) where the quartz phase was low, adding Ca-bentonite resulted in an increase in the bulk density, and for samples 9 and 11, compressive strength decreased remarkably.

The range of water absorption for these aggregates is 0–30% (Table 4). Some of these samples, such as the blue and the grey, had very low water absorption because the aggregates were covered with a glazed surface which formed from the amorphous contents, and such a surface affects the measured water absorption.

Plotting the *MHE* as a function of density (Fig. 7a and b), the results of the samples without bentonite do not show a statistical correlation ($R^2=0.21$), but a correlation ($R^2=0.77$) can be assumed when bentonite is added.

Comparing our results to studies (Table 1) where artificial additives were applied, it can be said that bentonite as natural additive is also suitable for enhancing the properties of LWAs.

Mineral phases and microstructure of lightweight aggregates

To define the mineral phases in the LWAs, 12 samples were selected. Heat treatment (Hungarian clays at 1225 °C and Egyptian clays at 1275 °C) of the clay aggregates resulted in the amorphous phase being the main phase in the samples. The mineral phases for the additive free aggregates were the same as the crystalline phases of the aggregates with Ca bentonite.

Besides the 66–78% glassy phase, some crystalline phases were also found. In the Hungarian samples, quartz, hematite, and mullite were the main phases (Table 5). In most of the Egyptian samples, mullite was the main crystalline phase, thanks to the significant kaolinite content. Besides quartz and hematite and feldspars, other minerals like cordierite, cristobalite, and tridymite were also found.

Iron-containing phases or pure iron powder play an important role in the expansion mechanism of the clays and affect the properties of aggregates (Bernhardt et al. 2014). As the iron oxide content of Ca-bentonite was relatively high (14.5%), this favoured the formation of hematite. Figure 8 shows the change in the amount of hematite for samples with and without additives. With the exception of samples 2 and 9, it was higher in all cases where bentonite was used.

Another factor that may affect the properties of the aggregates is the amorphous phase. The amount of amorphous content was much higher compared to the crystalline phase. In the case of clay ceramics, the increasing amorphous content with increasing temperature generally increases the density, as it is accompanied by shrinkage. This usually means

Fig. 6 **a** Bar chart of bulk density of aggregates with and without Ca-bentonite. **b** Bar chart of compressive strength of aggregates with and without Ca-bentonite

Fig. 7 **a** Relation between the bulk density and maximum height expansion without bentonite. **b** Relation between the bulk density and maximum height expansion with 10% of bentonite

Table 5 Crystalline phase of clay- Ca bentonite aggregates

Samples	B	G	Y	1	2	5	6	8	9	10	11	12
Amorphous, %	78	73.8	76.9	75.2	74.3	77.4	68.5	70.4	72.5	70.2	66.4	68
Crystallinity, %	22	26.2	23.1	24.8	25.7	22.6	31.5	29.6	27.5	29.8	33.6	32
Crystalline phase												
Cristobalite	4.8	6.3	4.3	5.3	5.1	3.7	5.6	4.7	4.3	4.8	5.8	12.4
Hematite	11.7	11	12.2	6.4	11.3	14.5	9.4	18.4	15.1	9.6	14.6	14
Microcline	5.8	12	7.2	8.1	9.2	10.9	3.7	2.3	3.7	9.7	3.6	4.5
Quartz	13.9	16.7	26.7	6.3	13	33.5	19.4	4.5	8.7	14.6	2.6	20.4
Anorthite	20	9	4.2	32.5	7.6	5.4	10.7	9	4.4	14.2	7	2.7
Cordierite	2	1.8	0	2	1	0.9	2.6	3	1	2.9	2.6	2.6
Mullite	31	32.7	32.4	35.4	38.6	23.7	41.9	53	56.9	36.4	58	33.6
Tridymite	10.8	10.5	13	4	14.2	7.4	6.7	5.1	5.9	7.8	5.8	9.8

Fig. 8 a Bar chart for the hematite content before and after 10% of bentonite addition **Table 6** Correlation analysis for the parameters of the statistical model

Parameters	MHE, %	BD, g/cm ³	UCS, MPa	Amorphous contents, %
MHE, %	1			
BD, g/cm ³	−0.77	1		
UCS, MPa	−0.25	0.48	1	
Amorphous contents, %	0.64	−0.80	−0.49	1

a more compact structure. However, in the case of LWAs, the pressure of the gas phase at the firing temperature and the amorphous phase in the plastic state together result in

an increase in the volume of the samples. Using correlation analysis, we can search for correlations between the properties of LWAs and amorphous content. Table 6 shows a correlation model (MS Excel) for the clays studied. From the correlation for regression values, the amorphous glassy contents have a good reciprocal relationship with the bulk density, with regression values of -0.8 , and there is a direct relationship with the height of expansion. Regarding the latter, the regression coefficient is only 0.64 , and this means it does not have a strong effect.

In order to observe the pore distribution and the microstructure of the LWAs, we prepared and analysed aggregates with SEM. The structure of LWA is consistent with the phase composition in all of the samples. The structure of Sample

Fig. 9 SEM micrograph of LWA at 1275 °C (a) clay sample (1) without bentonite ($M = \times 20$) (b) clay sample (1) with 10% of bentonite ($M = \times 50$)

Fig. 10 SEM micrograph of LWA at 1275 °C a clay sample (1) with 10 wt% of bentonite with the EDAX analysis to point 3 ($M = \times 100$). b Clay sample (5) with 10 wt% of bentonite with the EDAX analysis to point 5 ($M = \times 100$)

1 can be seen in Figs. 9 and 10 before and after the addition of bentonite. It can be seen in Fig. 9a that the cross-section of the aggregates consisted of two layers. Inside the outer layer, there are small pores with an irregular distribution. In contrast to this, the inner side is one large pore. This type of structure develops if the outer layer of the aggregates is closed by a silicate melt, and the evolved gas trapped inside and the amount of it that reaches the outer layer is very little. The process produces higher pressure inside the aggregate, so *MHE* increases and the density decreases.

After the addition of 10 wt% of bentonite, the structure of the sample was different. It can be observed in Fig. 9b that the pore distribution became homogeneous; the pore size distribution seems to be identical throughout the cross-section and the surface of aggregates is also smoother. The

new structure with bentonite is better, because regular pore size can increase the compressive strength of LWA, as it causes a more homogeneous stress distribution throughout the microstructure (Bernhardt et al. 2014; Cheeseman and Viridi 2005).

SEM images showed large amounts of white spots on the fracture surfaces of the samples, and these multiplied around the pores and on the surface of the pores. Figures 10a and b provide an example. EDAX studies have revealed that these are high iron phases. In the pictures, numbers indicate the studied areas; the chemical compositions of which are given in Table 7. Point 1, measured by EDAX, can be found in Fig. 9a.

Table 7 EDAX results of the clay- Ca bentonite aggregates

Samples		Point	C,%	O,%	Na,%	Mg,%	Al,%	Si,%	K,%	Ca,%	Ti,%	Fe,%
Additive free	1	1	5.04	30.44	0.12	0	0.21	62.88	0.26	0.29	0.19	0.56
Clay + 10 wt% of Ca-bentonite	1	2	2.66	23.78	2.2	0.86	12.8	47.89	4.57	1.34	0.37	3.52
		3	1.26	10.34	0.74	1.61	8.64	9.51	0.9	0.84	3.13	63.02
	5	4	2.71	21.8	1.49	1.89	11.71	43.76	2.54	6.4	1.55	6.16
		5	1.14	17.71	0.1	0.84	4.71	3.71	0.29	1.41	2.7	67.39

Conclusion

In contrast to previous research (Table 1) which used artificial additives to modify the properties of the expandable clays, in this study, a natural additive, bentonite, was used. The effect of Ca-bentonite as a natural additive on the properties of light expanded clay aggregate was studied. The addition of bentonite is intended to improve the properties of LWA and reduce the energy required for production. The behaviour of twelve clay-bentonite mixtures (three Hungarian and nine Egyptian clay samples) was examined.

Bentonite as an additive with a high iron oxide and clay mineral content clearly influenced and changed the properties of each clay. With the addition of bentonite, not only was more iron added to the mixture, but the decomposition of the introduced clay minerals resulted in additional alkali and alkaline earth metal oxides, which lowered the expansion temperature of the clay mixtures.

In the case of the Hungarian clays, the densities decreased, and the strength values doubled in some samples. Bentonite allowed for the production of much better quality LWA at lower temperatures for these clays. In addition to the economic benefits of lower combustion temperatures, CO₂ emissions can also be reduced.

In the case of the Egyptian clays, bentonite did not improve the properties in all cases. Of the nine Egyptian clays tested, five showed better sample strength (2–180% improvement), while four had a significant decrease (23–70% reduction). From the measured data, it can be seen that above 25% kaolinite content, in the case of Egyptian clays, bentonite addition increased the density of the samples, while at 20% lower kaolinite content, the density clearly decreased and the strength increased.

Not all clay is affected in the same way by the use of bentonite, as of course the constituent minerals of the clay also affect the results. It can be said that it is possible to improve the properties of LWAs with a natural additive like bentonite, but the appropriate amount must be selected for each clay and the sintering temperature of the mixture must be determined.

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Declarations

Conflict of interest The authors declare no competing interests.

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