ANTAL KERPELY DOCTORAL SCHOOL OF
MATERIALS SCIENCE & TECHNOLOGY

Production of Lightweight Concrete: Modeling and Assessment of Changes in Textures and Mineral Phases

Thesis Booklet

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1. Introduction

Lightweight concretes (*LWCs*) are promising structural materials nowadays. They can produce structures, bridges, and buildings with reduced density. The essential component of LWCs is the lightweight aggregate (*LWAs*), which can divide into the following groups: 1) natural aggregates, 2) industrial waste lightweight aggregates, and 3) artificial lightweight aggregates [1].

Lightweight aggregates can be processed or produced from porous stone, fly ash, expanded slag, volcanic scoria, perlite, and clay. The LWA is also an ideal primary material for LWCs products (*masonry blocks, slab elements, etc.*), lightweight load-bearing concrete, backfill, green roofs, and soil for ornamental plants and vegetable gardening.

Due to their thermal performance, sound insulation, and good fire resistance, lightweight aggregates are becoming the focus of interest worldwide [1, 2, 3]. Lightweight aggregates are made from clay, shale, slag, natural pumice, tuff, and perlite. The prepared LWAs from expanded clay must expand to approximately twice their original volume [3]. According to the definition, lightweight aggregates have the following physical and mechanical properties: a bulk density of less than 1.2 g/cm^3 and a particle density of less than 2 g/cm^3 [4, 5], and the uniaxial compressive strength exceeds 1 MPa [6]. In addition, lightweight expandable clay aggregates are porous ceramic products with a highly uniform pore structure [3, 7]. Besides, the aggregate shape is spherical [3]

During the industrial process, clay is crushed, screened to size, and fired in a rotary kiln. The LWA manufacturing process is divided into three major stages: pellet formation, sintering (firing), and defining the properties of these LWA. In the first stage, specific proportions of clay materials are grounded. The unfired granules can be formed by pelletizer or by hand with a suitable water addition [8, 9]. These granules are typically dried and sintered after granulation. The typical sintering temperatures for LWA are between 1000°C and 1300°C [10].

Sintering is the second stage for processing the LWA. With sintering, the most critical factors in the processing of LWA production are pyro-plastic deformation, gas generation, and gas retention. All these factors can be controlled in aggregate expansion (*bloating*).

The mechanism of expansion in LWAs, which takes place in several steps, was described by Riley [11]. The first step is the evolution of gasses at a specific temperature where the material is in its visco-plastic state. The gas generation sources in this process can be the reduction of ferric oxide, the combustion of organic matter, or the thermal decomposition of carbonates [12, 13, 14]. In the second step, a partially molten state of the clay prevents the escape of gases. Finally, after cooling, the molten phases remain as amorphous content containing different crystalline compounds depending on the composition of the raw material [9].

When the temperature is too high during sintering, excess gas can evolve through the surface of the aggregates, improving the overall porosity and creating continuous pores [15, 16].

In addition, one of the essential materials in building and construction is lightweight structural concrete (*LWC*). The advantages of the LWCs are a higher strength-to-weight ratio, high tensile strength, low thermal expansion, and good insulation properties, these advantages can be observed due to the air pores in the lightweight aggregate [17, 18].

2. Summary of literature

The literature review aims to show and introduce the theoretical background on the chemical and mineral composition of the expanded clays and its effect on LWA expansion. The properties of the LWA and LWC and the effect of their additives were also mentioned. Finally, the interfacial transition zone (*ITZ*), which affects the properties of the concrete cubes, is being studied.

The chemical composition of the expanded clay is the most crucial factor affecting the bloating mechanism. This mechanism is determined by the ratio of three chemical oxide groups: SiO_2, Al_2O_3, and total flux ($Fe_2O_3 + MgO + CaO + Na_2O + K_2O$) [11, 16].

Riley [11] proposed that bloating factors are.

1- The raw materials have a chemical composition that allows for the formation of amorphous content.
2- Gases are produced and trapped by the liquid phase formed by high temperatures.

Riley's study includes a bloating area where bloating behaviour occurs in a ternary diagram composed of SiO_2, Al_2O_3, and total flux.

The mineral composition is the second factor that influences the properties of LWA. During sintering, the mineralogy of the LWA changes. The mineralogical composition of the expanded clay deposits includes smectite, kaolinite, chlorite, and illite, as well as quartz, feldspars, and calcite. On the other hand, some mineral phases, such as muscovite and vermiculite, are associated with clay minerals. Many clay mineral deposits are able to bloat and can be used to form LWA [6, 19].

Additive materials can enhance the physical, mechanical, or expansion properties of the lightweight aggregates [20]. Some additives, such as iron, can improve expansion, but bulk density and uniaxial compressive strength are simultaneously reduced [21]. Generally, adding iron contents to LWA generated large pores in the middle part of the pellet but did not influence the strength of the aggregates, density, or total porosity [21].

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 [22].

The literature review in Table 1 shows the changes in the properties of the LWA with additive materials.

Table 1 The influence of the additive materials on the properties of LWAs

Author(s)	Additive types	Amount, [wt%]	Effect
Moreno-Maroto. et al., [2017] [23]	Carbon fibers	5-10	• Drop in particle density, • Increase in total porosity, • Enhanced compressive strength, • Carbon fiber can achieve energy savings.
Bernhardt. et al, [2014] [21].	Na_2CO_3,	2	• High open porosity with poor expansion.
	SiO_2,	5	• Increase in porosity.
	Fe_2O_3 and Fe	5	• Enhanced porosity and a high expansion.
Lee and Liu [2009] [24]	CaF_2	10–20	• Great influence on compressive strength.
Wei. et al., [2018] [22]	CaF_2	10–20	• Temperature decreases by 200°C.
Islam et al., [2016] [25]	Soda-lime silica glass	20-50	• Bulk density and water absorption decreased, • Improvement in crushing resistance of LWA.
Lee, [2016] [14]	Carbon and kerosene	2-4-6	• Decrease in density, • Lower expanding and sintering temperature.
Liu. et al., [2017] [26]	Autoplastic	1	• Increased porosity by 39.5%, • Bulk density decreased by 28.5%.
Fakhfakh et al., [2007] [27]	Quartz automobile oil	15+1	• High gas, Drop in bloating temperature. • High water absorption capacity.
Abdelfattah, et al., [2020] [28]	Bentonite	5-10-20	• Decreased in density, • High compressive strength, • High expansion

3. Knowledge gap and the aims of the thesis

Based on the literature review, most studies deal with the production and characterization of LWAs and LWCs; another group of articles deals with the modification of the properties of LWAs by artificial additives. There is no integrated model that predicts the properties (*bulk density and uniaxial compressive strength*) of the LWAs based on the characterization of the clays (*chemical, mineral analysis, and expansion data*). The effects of natural additives on lightweight clay aggregates and on the physical, mechanical and dilatation properties of concrete have also not been investigated.

Therefore, from the discussion above, some questions must be answered:

- Are there any advantages to predict the properties of the LWA?
- Is it possible to enhance the properties of LWAs by using natural additive materials?
- What kind of natural additive materials can be used with LWAs?

- What kind of phases can be developed after firing? In addition, which of these phases can be affected the properties of the *LWAs*?

The questions mentioned above determine the goals of my thesis:

- Examination of expanded clays as a raw material used in the sample preparation of LWAs.
- Use additive materials such as volcanic rocks (*tuff and perlite*) and sedimentary rock (*bentonite*) to improve the properties of LWA (*such as bulk and particle density, uniaxial compressive strength, and expansion*).
- Providing information and studying the development of mineral phases and their effect on the properties of LWAs.
- Developing an integrated statistical model to predict the properties (*bulk density and uniaxial compressive strength*) based on chemical oxides and expansion data.
- Finally, producing lightweight concrete using lightweight aggregates with some additive materials to improve the properties of LWC.

4. Materials and methods

Fifteen raw clay materials were collected from the Mályi quarry, Hungary (labelled Yellow (*Y*), Grey (*G*), and Blue (*B*)), as well as the quarries from Giza (*1*), Fayum (*2*), Marsa-matrouh (*3*), El- Alamin (*4*), Sinai (*5*), Helwan (*6, 7*), and El-Saf (*8, 9, 10, 11, 12*) in Egypt. Besides, three different additive materials (*perlite, rhyolite tuff, and bentonite*) were used as natural additives to modify the properties of the produced LWAs. The additive materials were also collected from Hungary. The perlite mineral deposit was collected near *Cser-hegy*, while the rhyolite tuff mine is near *Bodrogkeresztúr*. The third additive material was bentonite. The bentonite originated from a deposit in Hungary (*Istenmezeje*).

4.1. Samples preparation and testing methods

Clays were dried for 24 hours at 105°C. They were easily ground for 15 minutes at 200 rpm on a *Retsch PM 400 planetary* ball mill to reduce particle agglomeration and milled to pass through a 100 µm opening sieve for further measurement [2, 20].

All major, minor oxides and trace elements in raw clay materials were characterized using X-ray fluorescence (*XRF*) on Rigaku Supermini 200 WDXRF instrument.

The mineralogical analysis for raw clay materials and lightweight and crushed lightweight concrete was examined using the X-ray diffraction (*XRD*) reference *Rigaku* Miniflex *II.* The phase identification and quantitative Rietveld refinements were achieved using the Profex 4.3 software.

The thermal characterization, comprising concurrent differential and thermogravimetric studies (*TG/DTA*), was measured by *MOM Derivatograph-C* instrument. A heating microscope (*Camar Elettronica*) was used to observe the sintering, melting properties, and dimensional changes of the raw clay based on equation (1)

$$HE = (L - L_0) / L_0 \cdot 100\ [\%] \qquad (1)$$

Its maximum value is the maximum height expansion (*MHE*).

After the analysis of raw materials, the green pellets were made by hand rolling and using by handmade pelletizer. The diameter of the green pellets ranged from 6 to 14 mm were used to create the LWAs. The processing of LWAs is summarized in Figure 1.

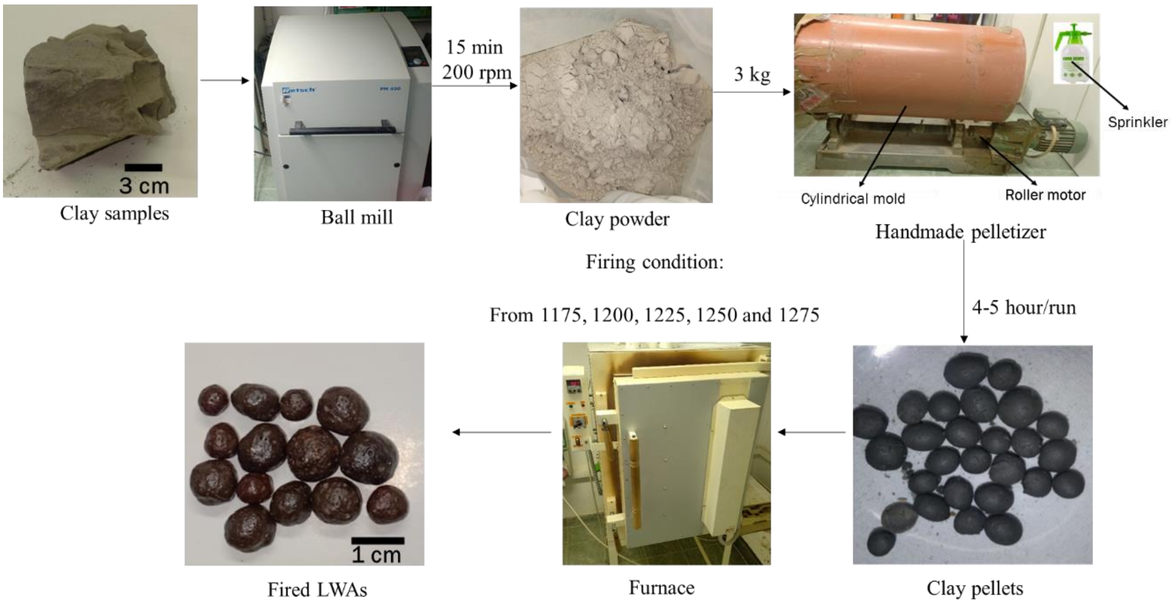


Figure 1 Processing of the LWAs

The pellets were dried in a drying chamber at 105°C. This method was used for clay without additive *(additive-free*) materials. Then the clays were mixed with 10 wt% of perlite, tuff, and bentonite materials.

Dried pellets were sintered in an air atmosphere using an electrical furnace for 10 minutes at temperatures ranging from 1150°C to 1275°C with a heating/cooling rate of 10°C/min. In this study, the beneficial properties (*such as bulk density, particle density, and uniaxial compressive strength*) of the samples were achieved at 1225°C for Hungarian and 1275°C for Egyptian samples. Lastly, sintered products were naturally cooled down in the furnace after heating.

The bulk density and water absorption, and uniaxial compressive strength (*UCS*) of the aggregates were measured and analysed.

4.2. Modelling

I try to build an integrated model using various physical, mechanical, and mineralogical data from the LWAs. All the data extracted from my previous experiments were collected in one multilinear regression model. Three significant steps must be taken to implement this model: correlation, ANOVA (analysis of variance) test, and multilinear regression.

4.3. Concrete mixing batches

Following the firing, the fired pellets were mixed with cement and water to create concrete. The cement used was ordinary Portland cement (*OPC*), with a uniaxial compressive strength of 42.5 MPa after 28 days. The concrete components ratio was 1:2:4 (*water:cement: aggregates*) with aggregates size from (*4.0-8.0 mm*), with a water/cement ratio of 0.5. After one day of molding, the concrete cubes were removed from the mold and placed in a water tank for 28 days of curing. The cubes were cured following the identification of the standard [29]. After 28 days, the bulk density, water absorption, specific gravity, and uniaxial compressive strength of the concrete samples were measured. Finally, the microstructure, surface morphology, and pore size of the different LWAs and LWCs were examined by scanning electron microscope (*SEM*) with an *EDAX* unit attached to the SEM microscope.

5. Results and discussion

5.1. Materials characterization

After the chemical analysis of the raw materials, the main oxide groups (*silica, alumina, and total flux*) that could affect bloating process were determined. Concerning the Riley diagram [11], all 15 clays were in the bloating area (*red contour area*), which means that these clays can expand/bloat upon firing below 1300°C [30], as exhibited in Figure 2.

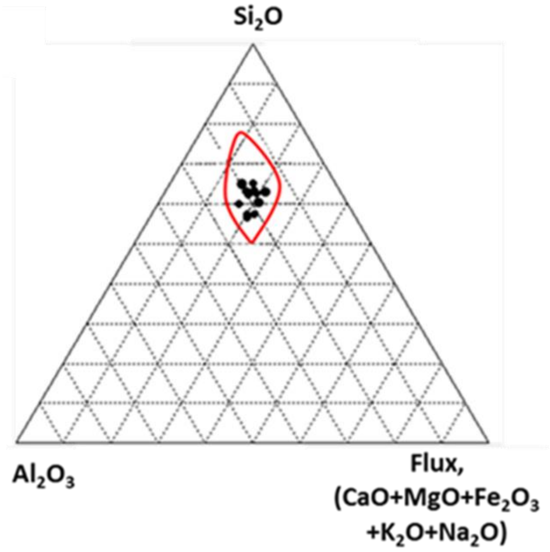


Figure 2. Chemical composition of the technological sample as plotted on Riley's diagram (1951)

XRD analysis revealed that the main constituents in the clays are quartz, illite, montmorillonite, kaolinite, calcite, albite, and smectite. A heating microscope was used to characterize the melting and bloating behaviour of the raw clay materials samples. Table 2 shows the maximum height expansion for the clay samples.

Table 2 Maximum height expansion (*MHE*) of additive-free clay

Sample	**Hungarian**			**Egyptian**											
	B	**Y**	**G**	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
MHE [%]	138	122	123	129	122	108	120	122	122	118	126	112	144	123	121

The highest height expansion of the Blue, Grey and Yellow clay samples were 138%, 123%, and 122%, respectively. At the same time, the Egyptian raw clay materials ranged from 108 to 144 % (*Table 2).* As shown in Figure 3a, the height expansion of the Hungarian clay samples improved as the temperature raised to a specific temperature, after which it began to drop to melt the material.

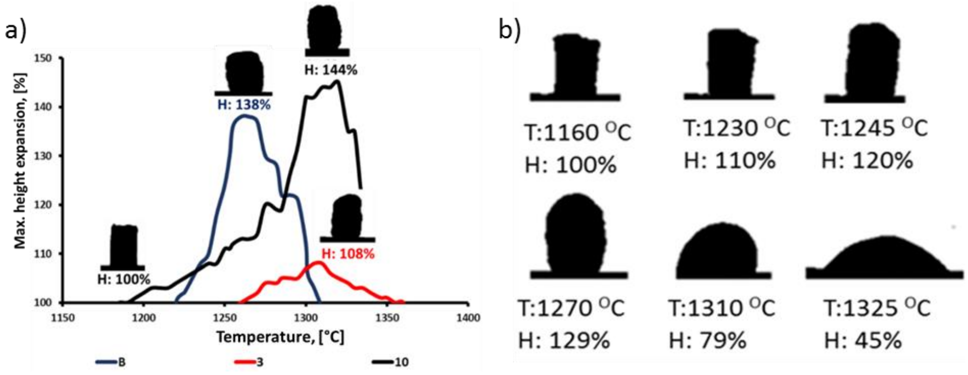
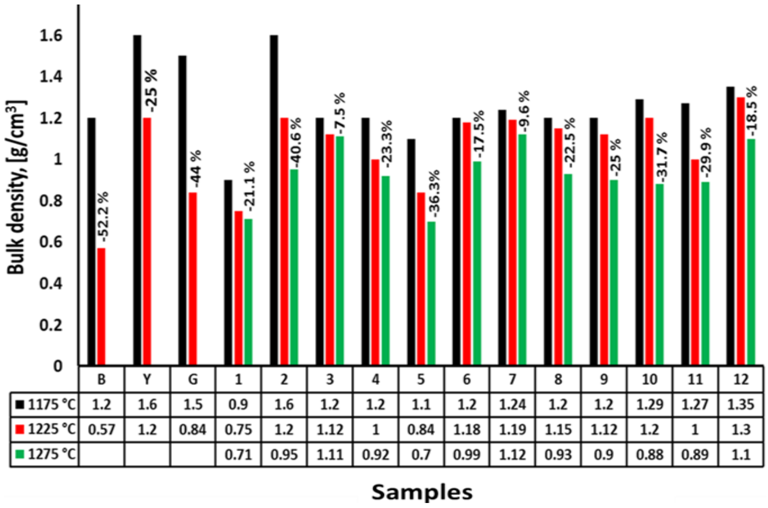


Figure 3 a) Relationship between height expansions of clay samples with the temperature, b) the height expansion of clay Sample 1 without additive at different temperatures

Image analysis revealed the characteristic temperatures (*T*) and actual height (*H*) of the sample, shown in Figure 3b.

5.2. Characterization of LWAs without additive materials

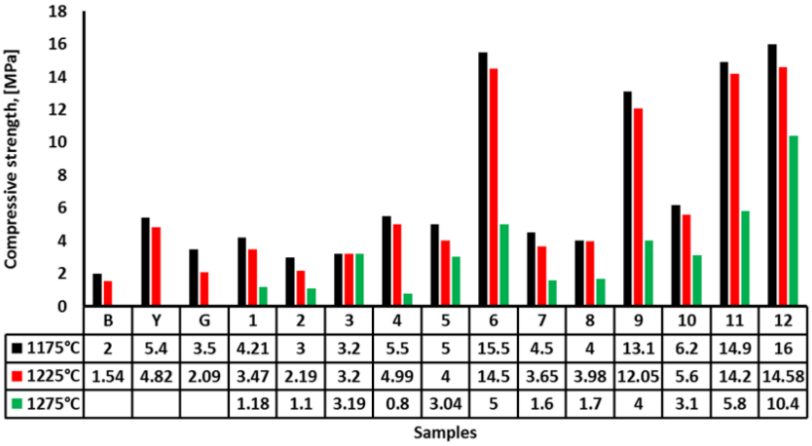
The results of the density measurements of the 15 different samples fired at different temperatures were summarized in Figure 4. The first observation was that the densities of all clays and clay mixtures tested were below (*or equal*) 1.2 g/cm^3. All the LWAs produced met the requirements for lightweight aggregates according to EN 13055-1: (2002). The numbers in the bar graph show the rate of decrease in density relative to the lowest firing temperature.



	B	Y	G	1	2	3	4	5	6	7	8	9	10	11	12
■ 1175 °C	1.2	1.6	1.5	0.9	1.6	1.2	1.2	1.1	1.2	1.24	1.2	1.2	1.29	1.27	1.35
■ 1225 °C	0.57	1.2	0.84	0.75	1.2	1.12	1	0.84	1.18	1.19	1.15	1.12	1.2	1	1.3
■ 1275 °C				0.71	0.95	1.11	0.92	0.7	0.99	1.12	0.93	0.9	0.88	0.89	1.1

Figure 4 Bar chart of bulk density of expandable clay aggregates

Besides bulk density, the uniaxial compressive strength of the LWAs is another crucial parameter for their application. According to the test results, the uniaxial compressive strength of the aggregates decreased with increasing temperature, as shown in Figure 5. The uniaxial compressive strength of all the LWAs at the firing temperatures was higher than the required limit (*1.0 MPa*), except for Sample 4 (*UCS= 0.8 MPa*). The uniaxial compressive strength values of the fired LWAs without additives were between 1.54–4.82 MPa for the Hungarian aggregates (*fired at 1225°C*) and were in the range of 1.1–10.4 MPa for the Egyptian aggregates (*fired at 1275°C*).



	B	Y	G	1	2	3	4	5	6	7	8	9	10	11	12
■ 1175°C	2	5.4	3.5	4.21	3	3.2	5.5	5	15.5	4.5	4	13.1	6.2	14.9	16
🟥 1225°C	1.54	4.82	2.09	3.47	2.19	3.2	4.99	4	14.5	3.65	3.98	12.05	5.6	14.2	14.58
🟩 1275°C				1.18	1.1	3.19	0.8	3.04	5	1.6	1.7	4	3.1	5.8	10.4

Figure 5 Bar chart of UCS of expandable clay aggregates

5.3. The mineralogical modification on the Riley diagram

Based on an analysis of the mineral and oxide composition of expanded clays from different deposits and the LWAs produced using the clays, I modified the Riley diagram used to determine the expandability of clays. The idea of the modification is that instead of SiO_2 content, the quartz content of the sample is plotted alongside Al_2O_3 and Flux after normalizing each value, as shown in Figure 6. In the modified Riley diagram, the samples can be sorted by quartz content based on the experiments performed. The highest quartz content (*horizontal bold black line in Figure 6*), which is, in this case, the limit (*32.5 wt% for the Yellow sample*) for bulk density of aggregates of 1.2 g/cm^3. It can be stated that below this quartz content, all samples have a density that meets the European standard.

Based on the analysis of the samples, an area marked with a green elliptic can also be identified in the diagram where the density of LWAs is less than 1.0 g/cm^3. Within this area, the density of the samples (*marked in a black number*) is below 1.0 g/cm^3. Samples Yellow and 7, 3, and 12 are outside this area. These are indicated with red numbers.

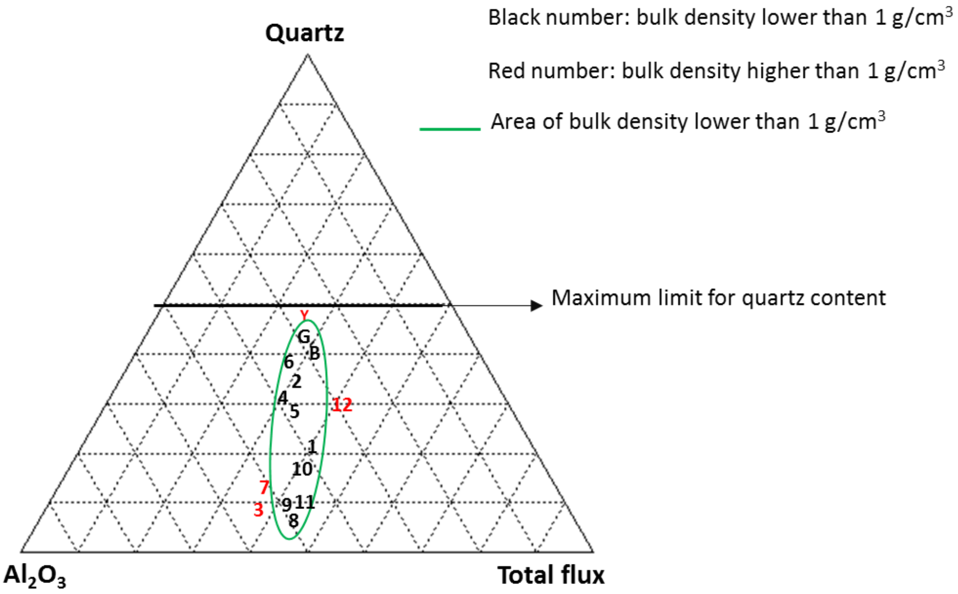


Figure 6 Ternary diagram between quartz, Al_2O_3, and flux oxides (*Modifying on Riley Diagram*)

5.4. The statistical models of LWAs

All the data extracted from previous experiments were collected in one statistical model, which is expected to predict the bulk density or uniaxial compressive strength of the LWAs by a multilinear regression model without measuring it. To integrate statistical correlation into one model, three different steps are necessary to take. The steps are as follows: 1) correlation, 2) ANOVA, and 3) multilinear regression.

The bulk density, UCS, MHE, and the three main oxide groups (*SiO_2, Al_2O_3, total flux*) of the LWAs were the most critical factors for building this model. The research aimed to produce LWAs, samples that meet the standard of density less than 1.2 g/cm^3 and uniaxial compressive strength greater than 1 MPa. Therefore, all used samples in these two models having a bulk density of less than 1.2 g/cm^3 with uniaxial compressive strength of more than 1 MPa were recorded in these models and calculated only by 14 samples. Two statistical tests can derive a statistical equation that can calculate the bulk density and uniaxial compressive strength of the LWAs. The ANOVA test considers the measured average of all parameters compared with the bulk density and uniaxial compressive strength. The multilinear regression model is the third step for this model that can be applied to these data. The third step of the model is multilinear regression. In this step, the relationship between two or more independent variables and a dependent variable is determined, and predictions and calculations are made for the dependent variable.

From the model, two equations can be determined to predict the bulk density (*BD*) (2) and the uniaxial compressive strength (*UCS*) (3) as follows:

$$\mathbf{BD = 13.084- (0.008* MHE) -(0.117* SiO_2)– (0.126* Al_2O_3) -(0.134* FX) (25\ \%\pm)} \quad (2)$$

$$\mathbf{UCS = 187.882-(0.197*MHE) -(1.705*SiO_2)–(2.838*Al_2O_3) -(1.177*FX) (220\ \%\pm)} \quad (3)$$

5.5. Discussion of the properties of additive-clay aggregates

The addition of natural additives affected each clay sample studied from different deposits. Additives have modified the properties of the manufactured LWAs compared to clays without additives. Table 3 shows the values and the effect of these additives on the maximum height expansion (*MHE*), bulk density, and uniaxial compressive strength of LWAs. The effects of perlite, tuff, and bentonite are shown separately in Table 3, indicated only by symbols, excluding the degree of change. Those cells that contain an improved property of the given LWA are highlighted in green.

The lightweight additives (*LWA*) should be as lightweight and strong as possible; therefore, they can be used to build lighter but stronger concrete structures. Natural additives are the most effective when the bulk density of the lightweight aggregate produced decreases and its strength grows simultaneously. This effect was achieved for 8 samples (*B, Y, G, 2, 4, 5, 6, and 7*) with 10 wt% of perlite, 5 samples (*B, Y, G, 2, and 7*) with tuff, and 6 for bentonite (*B, Y, G, 2, 7 and 12*).

Although the chemical composition and amorphous content of perlite and rhyolite tuff are nearly identical, the two additives have similarly altered the properties of LWAs made from younger Pannonian clays. However, the effect was not in the same direction for the older and higher kaolinitic clays of the middle Eocene to late Pliocene age and the oldest Senonian age.

Egyptian clays have a significantly higher kaolin content, which influenced both the firing temperature and the effect of additives.

Table 3 The properties and changes of LWAs after 10 wt% addition of perlite, tuff, and bentonite

Samples	B	Y	G	1	2	3	4	5	6	7	8	9	10	11	12
Properties of LWAs with perlite addition															
MHE, %	153	133	134	143	137	110	118	137	132	117	126	128	122	124	121
BD, g/cm^3	0.53	0.96	0.82	0.7	0.94	1.08	0.91	0.63	0.98	1.1	0.92	0.99	0.92	0.82	0.99
UCS, MPa	1.8	5.73	2.5	1.18	1.7	3.2	1	3.9	6.2	5.58	0.75	3.01	3.1	5.5	8.91
Properties of LWAs with tuff addition															
MHE, %	157	125	139	130	127	109	122	144	130	126	133	117	121	120	118
BD, g/cm^3	0.54	1	0.6	0.64	0.92	1.13	0.93	0.66	0.99	1.03	0.95	0.92	1	0.95	1.05
UCS, MPa	2.69	5	3	1.06	5.08	0.89	0.9	1.4	7.2	2.96	0.65	5.2	3.2	4.8	3.2
Properties of LWAs with bentonite addition															
MHE, %	164	135	133	135	129	101	116	135	124	112	123	132	120	133	124
BD, g/cm^3	0.55	0.95	0.79	0.75	0.94	1.1	1	0.65	0.95	1.1	0.97	0.99	0.9	0.98	1.02
UCS, MPa	3.27	7.15	4.32	1.2	2	3	0.8	1.46	4.2	1.86	4.78	1.2	3.8	4.78	12.08
Effect of the additive materials on LWAs properties															
Change with perlite addition															
MHE, %	↑	↑	↑	↑	↑	↑	↓	↑	↑	↓	≡	↑	↓	↑	≡
BD, g/cm^3	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	↓	↑	↑	↓	↓
UCS, MPa	↑	↑	↑	≡	↑	≡	↑	↑	↑	↑	↓	↓	≡	↓	↓
Change with tuff addition															
MHE, %	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↑	↓	↓	↓
BD, g/cm^3	↓	↓	↓	↓	↓	↑	↑	↓	≡	↓	↑	↑	↑	↑	↓
UCS, MPa	↑	↑	↑	↓	↑	↓	↑	↓	↑	↑	↓	↑	↑	↓	↓
Change with bentonite addition															
MHE, %	↑	↑	↑	↑	↑	↓	↓	↑	↑	↓	↓	↑	↓	↑	↑
BD, g/cm^3	↓	↓	↓	↑	↓	↓	↑	↓	↓	↓	↑	↑	↑	↑	↓
UCS, MPa	↑	↑	↑	↑	↑	↓	↑	↓	↓	↑	↑	↓	↑	↓	↑

**BD: Bulk density; UCS: Uniaxial compressive strength; ↓: properties decreased; ↑: properties increased; ≡ no change green colour; positive influence, red colour; negative influence.*

5.5.1. Mineralogical characterization of the perlite, tuff, and bentonite-clay aggregates

The amorphous phase is naturally dominant in all the LWAs prepared from additive-free clay samples, ranging from 65-76 wt%, respectively. In the crystalline phase, in the clays containing higher amounts of kaolinite, mainly mullite can be found, but all samples contained varying amounts of quartz polymorphs (*quartz, tridymite, cristobalite*), hematite, and feldspars such as anorthite and microcline. Hematite indicates the presence of a gas-forming phase containing iron. The amorphous content increased slightly in the samples with 10 wt% perlite or tuff, as they contained a large amount of amorphous phase. The crystalline phases were the same as it was formed in additive-free samples.

Considering the mineral phases formed in all clays, a stronger coefficient of determination between the constituent phases and the physical properties of the LWAs was only found between the amount of amorphous phase and the bulk density of the samples, as shown in Figure 7.

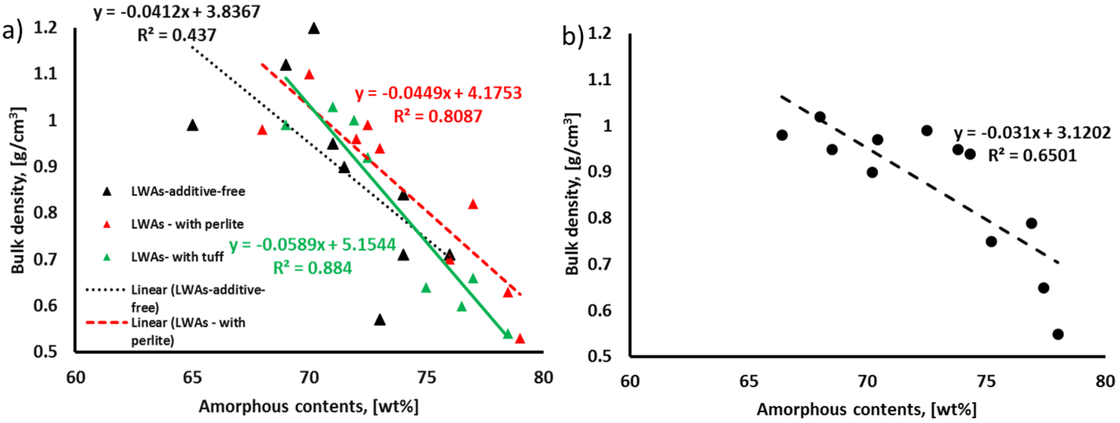
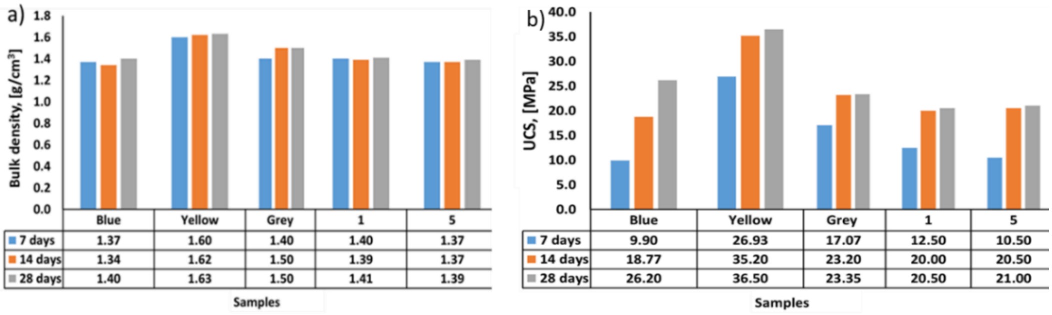


Figure 7 Trends between sample density and amorphous contents for a) perlite and tuff, b) bentonite

The strongest tendencies were observed for the perlite, tuff, and bentonite aggregates, where R^2 is 0.80, 0.88, and 0.65, respectively. The trend indicates that the huge amount of amorphous content is more effective in trapping evolving gases, reducing the density of the samples.

5.6. The properties of the LWC without and with additive materials

From all previous measurements on LWAs, I choose to represent 5 samples (*Blue, Yellow, Grey, 1, and 5*) for producing the concrete cubes. All previous experiments show that the bulk density and UCS of the additive-free concrete cubes at 28 days are less than 1.8 g /cm^3 (F*igure 8a*) and have a maximum strength higher than 17.0 MPa (*Standard limit*) (*Figure 8b*). According to the standards, these samples had values matched with the limits of bulk density and the UCS. As a result, these samples can be used as structural LWC.



	Blue	Yellow	Grey	1	5
🟦 7 days	1.37	1.60	1.40	1.40	1.37
🟧 14 days	1.34	1.62	1.50	1.39	1.37
⬜ 28 days	1.40	1.63	1.50	1.41	1.39

	Blue	Yellow	Grey	1	5
🟦 7 days	9.90	26.93	17.07	12.50	10.50
🟧 14 days	18.77	35.20	23.20	20.00	20.50
⬜ 28 days	26.20	36.50	23.35	20.50	21.00

Figure 8 a) Bulk density for the LWC, b) UCS for the LWC

Three different temperatures were used to investigate the effect of different thermal loads on the properties of LWCs with LWAs prepared without and with additives. As expected, the UCS decreased at higher temperatures (*300 °C and 500 °C*). However, at 200 °C, the measurement results showed a small decrease in bulk density caused by the dehydration process of the concrete structure. Parallel to this, an increase was observed in the UCS results.

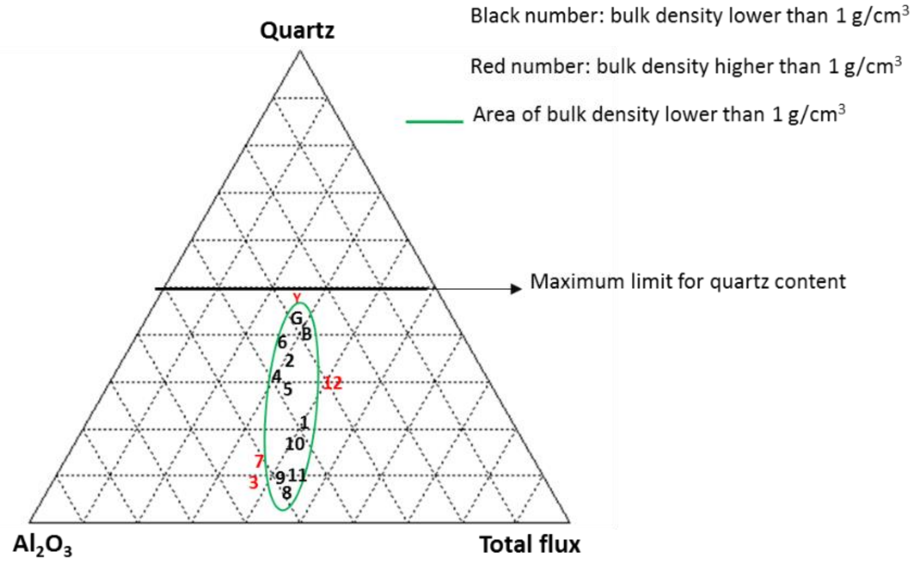
6. NEW SCIENTIFIC RESULTS

Thesis 1

A modification of the Riley diagram, when the normalized value of the amount of quartz phase and Al_2O_3 and fluxing oxides (Fe_2O_3 + MgO + CaO + Na_2O + K_2O) of an expandable clay sample are plotted, can be used to identify lightweight aggregates, fired at optimal temperature, with a bulk density less than 1 g/cm^3.

The modified Riley diagram is applicable for expandable clays for which:

- The optimal firing temperature is 1225°C for clays with low kaolinite content (*1-4 wt%*) and is 1275°C for clays with high kaolinite content (*13-34 wt%).*
- The quartz content of expandable clays is between 2.99-32.5wt%, Al_2O_3 content is between 17.29-22.53 wt%, and fluxing oxides content is between 15.97-21.45 wt%.



Ternary diagram between quartz, Al_2O_3, and flux oxides (*Modifying on Riley Diagram*)

Thesis 2

An integrated statistical model for expandable clays from different deposits, based on the SiO_2, Al_2O_3, fluxing oxides (Fe_2O_3 + MgO + CaO + Na_2O + K_2O) composition (*wt%*) and the maximum sample height (*%*) measured by a heating microscope, can be used to estimate the bulk density of lightweight aggregates by using the following multilinear regression equation:

Bulk density= 13.084- (0.008* Maximum height expansion) - (0.117* SiO_2) – (0.126* Al_2O_3) - (0.134* Fluxing oxide) (Error %±)

The integrated statistical model can be used for expandable clays with quartz contents varying between 55.15-64.97 wt%, Al_2O_3 contents varying between 17.29-22.53 wt%, and fluxing oxide contents varying between 15.97-21.45 wt%.

Thesis 3

For expandable clays where the kaolinite content is lower than 4 wt%, the application of 10 wt% additives of perlite (*Cser-hegy*) or rhyolite tuff (*Bodrogkeresztúr)* or bentonite (*Istenmezeje)* affect the expansion process positively, the maximum height expansion of the clays increases in the range of 16-69% which results in 3-28% lower lightweight aggregate bulk density and 3-112% higher uniaxial compressive strength compared to the clays without additives.

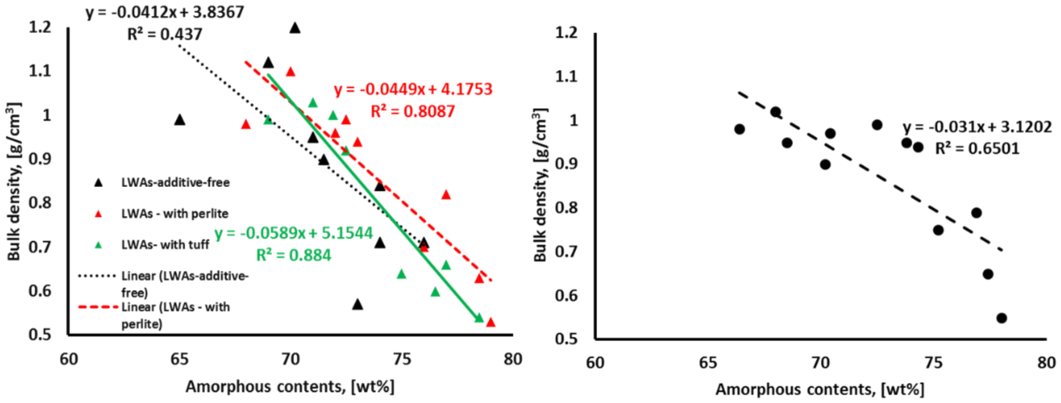
Thesis 4

The use of 10 wt% of perlite (*Cser-hegy*) or rhyolite tuff (*Bodrogkeresztúr*), or bentonite (*Istenmezeje*) as an additive for expandable clays alters the expansion process. It can cause a reduction in the bulk density of lightweight additives of about 8 to 20% for perlite, about 14 to 25% for tuff, and about 6 to 21% for bentonite and an increase in the uniaxial compressive strength of the lightweight aggregates between 14-250% for perlite, between 13-362% for tuff, and between 16-181% for bentonite.

Thesis 5

For lightweight aggregates, with a bulk density lower than 1.2 g/cm^3 and uniaxial compressive strength higher than 1.0 MPa, made of expandable clays from different deposits which used without or with additives such as perlite (*Cser-hegy*) or rhyolite tuff (*Bodrogkeresztú*r) or bentonite (*Istenmezeje*), the amorphous content (*in wt%)* of a lightweight aggregate is inversely proportional to the bulk density (*g/cm*^{3}). The inverse proportionality can be described by the following equations for samples with amorphous contents between 65-80 wt%:

a) for lightweight aggregates without additives: $y = -0.0412x + 3.8367$.
b) for perlite lightweight aggregates: $y = -0.0449x + 4.1753$.
c) for tuff lightweight aggregates: $y = -0.0589x + 5.1544$.
d) for bentonite lightweight aggregates: $y = -0.031x + 3.1202$.



Trends between sample density and amorphous content of the additives

7. LIST OF PUBLICATIONS

International Journal articles

Abdelfattah M, Géber R, Kocserha I.: Enhancing the properties of lightweight aggregates using volcanic rock additive materials. Journal of Building Engineering. (2022). V 63, Part A, 2023, 105426. https://doi.org/10.1016/j.jobe.2022.105426 (Q1, IF=7.144)

Abdelfattah M, Géber R, Abdel-Kader N A, Kocserha I. Assessment of the mineral phase and properties of clay-Ca bentonite lightweight aggregates. Arabian Journal of Geosciences (2022) 15:205 https://doi.org/10.1007/s12517-022-09538-w (Q2, IF=1.85)

Abdelfattah M, Kocserha I, Géber R, Tihtih M.: Evaluating the properties and mineral phases of the expanded clay aggregates with the bentonite additive material. Journal of Physics: Conference Series, 1527 (2020) 012030. doi:10.1088/1742-6596/1527/1/012030 (Q3)

Abdelfattah M, Kocserha I, Géber R. Effects of weathering grade of metavolcanic aggregates on mortars. IOP Conf. Series: Materials Science and Engineering 613 (2019) 012046. doi:10.1088/1757-899X/613/1/012046

Abdelfattah M, Géber R, Kocserha I.: Enhancement the properties of lightweight concrete mortars by some additive materials. Journal of Physics: Conference Series, 2315 (2022):012005. DOI: 10.1088/1742-6596/2315/1/012005

Tihtih M, Ibrahim J F M, Kurovics E, **Abdelfattah M.:** Study on the effect of Bi dopant on the structural and optical properties of $BaTiO_3$ nanoceramics synthesized via sol-gel method. Journal of Physics: Conference Series. 1527 (2020) 012043 doi:10.1088/1742-6596/1527/1/012043.

Conference Proceeding

Abdelfattah M, Kocserha I, Géber R. Effect of firing on mineral phases and properties of lightweight expanded clay aggregates. MultiScience - XXXIII. microCAD International Multidisciplinary Scientific Conference University of Miskolc, 23-24 May, 2019, ISBN 978-963-358-177-3

Abdelfattah M, Kocserha I, Géber R. The effect of calcium fluoride on mineral phases and properties of lightweight expanded clay aggregates. Conference: Proceedings – XIII th International Conference. Preparation of ceramic materials Jahodná, 25th–27th June, 2019, pp. 141–146. ISBN 978-80-553-3314-4 At: Slovakia

Oral presentation

Abdelfattah M. Effect of firing on mineral phases and properties of lightweight expanded clay aggregates. MultiScience - XXXIII. microCAD International Multidisciplinary Scientific Conference University of Miskolc, 23-24 May, 2019. Miskolc, Hungary.

Abdelfattah M. The effect of calcium fluoride on mineral phases and properties of lightweight expanded clay aggregates. Conference: Proceedings – XIII th International Conference. Preparation of ceramic materials Jahodná, 25th–27th June, 2019, pp. 141–146. At: Slovakia

Abdelfattah M. The effect of bentonite on the properties of lightweight expanded clay aggregates. International scientific conference on advanced in mechanical engineering. 7-9-November 2019. Debrecen, Hungary

Abdelfattah M. Integrated modeling for the properties and mineral phases of lightweight aggregates. The 2nd European Conference on Silicon and Silica Based Materials. October 2021. Miskolc-Lillafüred, Hungary

Abdelfattah M. Effect of tuff material on the properties of the lightweight expanded clay aggregates Miklós Iványi International PhD & DLA Symposium. 2020. Pecs, Hungary

Abdelfattah M. Effect of the additive materials on the properties of lightweight concrete aggregates. Miklós Iványi International PhD & DLA Symposium. October 2021. Pecs, Hungary

Poster Presentations

Abdelfattah M. Effect of the alkali silica reaction on metavolcanic mortars. The 5th International Conference on Competitive Materials and Technology Processes (ic-cmtp5). October 2018. Miskolc, Hungary.

Abdelfattah M. The effect of additive materials on phases and properties of lightweight expanded clay aggregates. The 1st European Conference on Silicon and Silica Based Materials. October 2019 Miskolc-Lillafüred, Hungary

Abdelfattah M. Effect of perlite material on the lightweight expanded clay. 6th International Conference on Competitive Materials and Technology Processes. October 2021, Miskolc-Lillafüred, Hungary.

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References

1. Choudhry V, Hadley S R, Utilization of Coal Gasification Slag: Overview. Praxis Engineers, Inc, Milpitas. (2009).
2. Liao Y, Huang C, Effects of heat treatment on the physical properties of lightweight aggregate from water reservoir sediment, Ceramics International. (2011). 37. p. 3723–3730. https://doi.org/10.1016/j.ceramint.2011.04.122
3. Rashad A M, Lightweight expanded clay aggregate as a building material – An Overview. Construction and Building Materials. (2018). 170. p. 757–775. https://doi.org/10.1016/j.conbuildmat.2018.03.009
4. EN 13055-1, Lightweight aggregates - Part 1: Lightweight aggregates for concrete, mortar and grout. (2002). https://doi.org/10.1016/j.conbuildmat.2017.12.018
5. Cheeseman C R, Virdi G S, Properties and microstructure of lightweight aggregate produced from sintered sewage sludge ash. Resour. Conserv. Recycl. (2005). 45, 18–30. https://doi.org/10.1016/j.resconrec.2004.12.006
6. Huang C H, Wang S Y, Application of water treatment sludge in the manufacturing of lightweight aggregate. Construction Building Material. (2013). 43, 174–183. https://doi.org/10.1016/j.conbuildmat.2013.02.016
7. González-Corrochano B, Alonso-Azcárate J, Rodríguez L, Lorenzo A P, Torío M F, Ramos J J T, Corvinos M D, Muro C, Valorization of washing aggregate sludge and sewage sludge for lightweight aggregates production, Construction Building Material. (2016). 116, 252–262. https://doi.org/10.1016/j.conbuildmat.2016.04.095
8. Alexander MG, Arliguie G, Ballivy G, Bentur A, Marchand J, Engineering and Transport Properties of the Interfacial Transition Zone in Cementitious Composites. RILEM Publications. (1999). ISBN: 2-912143-08-X
9. Ozguven A, Ozguven L X, Examination of effective parameters for the production of expanded clay aggregate. Cement and Concrete Composite. (2012). 34. p. 781–787. https://doi.org/10.1016/j.cemconcomp.2012.02.007
10. Chandra S, Berntsson L, Lightweight Aggregate Concrete Microstructure. Science, Technology, and Applications, Chapter N_6, (2002) 131–164. https://doi.org/10.1016/B978-081551486-2.50009-2
11. Riley C M, Relationship of chemical properties to bloating of clays. Journal of American ceramics society. (1951). 34: 121–128. https://doi.org/10.1111/j.1151-2916.1951.tb11619.x
12. Rudnai, G., Lightweight concretes, Akadémiai Kiadó. Budapest. (1963).
13. Chopra S., Lal K., Ramachandran V, Gas-producing agents in the production of lightweight aggregates. J. Chem. Technol. Biotechnology. (1964). 14 (5), 181–185.
14. Lee, K G, Bloating Mechanism of Lightweight Aggregate with the Size. Journal of Korean Ceramics Society. (2016). 53 (2). https://doi.org/10.4191/kcers.2016.53.2.241
15. Yang C, Cuin C, Qin J, Recycling of low-silicon iron tailings in the production of lightweight aggregates. Ceramics International. (2015) 41: 1213–1221.http://dx.doi.org/10.1016/j.ceramint.2014.09.050
16. Dondi M, Cappelletti P, D'Amore M, de Gennaro R, Graziano S F, Langella A, Raimondo M, Zanelli C, Lightweight aggregates from waste materials: Reappraisal of expansion

behavior and prediction schemes for bloating. Construction and Building Materials. (2016) 127, 394–409. https://doi.org/10.1016/j.conbuildmat.2016.09.111

17. Topcu I B, Semi lightweight concretes produced by volcanic slags. Cement and concrete research. (1997). 27 15– 21. https://doi.org/10.1016/S0008-8846(96)00190-1
18. Al-Khaiat H, Haque M N, Effect of initial curing on early strength and physical properties of a lightweight concrete. Cement and concrete research. (1998). 28, 859– 866. https://doi.org/10.1016/S0008-8846(98)00051-9
19. Tang C W, Chen H J, Wang S Y, Spaulding J, Production of synthetic lightweight aggregate using reservoir sediments for concrete and masonry. Cement and Concrete Composite. (2011). 33, 292–300. https://doi.org/10.1016/j.cemconcomp.2010.10.008
20. Kocserha I, Gömze LA, Friction properties of clay compounds. Applied Clay Science. (2010). 48: 425-430. doi:10.1016/j.clay.2010.01.017
21. Bernhardt M, Justnes H, Tellesb H, Wiik K, The effect of additives on the properties of lightweight aggregates produced from clay. Cement and Concrete Composite. (2014). 53, 233–238. https://doi.org/10.1016/j.cemconcomp.2014.07.005
22. Wei Y, Weng S, Xie X, Reduction of sintering energy by application of calcium fluoride as flux in lightweight aggregate sintering, Construction and Building Materials. (2018). 190, 765–772. https://doi.org/10.1016/j.conbuildmat.2018.09.134
23. Moreno-Maroto JM, González-Corrochano B, Alonso-Azcárate J, Rodríguez L, & Acosta A, Manufacturing of lightweight aggregates with carbon fiber and mineral wastes. Cement and Concrete Composites. (2017). 83: 335 -348.
24. Lee, TC, & Liu, FJ, Recovery of hazardous semiconductor-industry sludge as a useful resource. Journal of Hazardous Materials. (2009). 165(1-3): 359–365. doi:10.1016/j.jhazmat.2008.10.105.
25. Islam M, Sagirul S N, Moniruzzaman M, Umme A US, Effect of soda lime silica glass waste on the basic properties of clay aggregate, Int. J. Sci. Eng. Res. (2016). 7 (4), 149–153. ISSN 2229-5518
26. Liu P, Farzana R, Rajarao R, Sahajwalla V, Lightweight expanded aggregates from the mixture of waste automotive plastics and clay, Construction and Building Materials. (2017). 145. 283–291.
27. Fakhfakh E, Hajjaji W, Medhioub M, Rocha F, López-Galindo A, Setti M, Kooli F, Zargouni F, Jamoussi F, Effects of sand addition on production of lightweight aggregates from Tunisian smectite-rich clayey rocks. Applied Clay Science. (2007). 35, 228–237. doi:10.1016/j.clay.2006.09.006
28. Abdelfattah M, Kocserha I, Géber R, Tihtih M, Móricz F, Evaluating the properties and mineral phases of the expanded clay aggregates with the bentonite additive material, Journal of Physics: Conference Series. (2020). 1527, 012030. doi:10.1088/1742-6596/1527/1/012030.
29. ASTM C330/C330M-09, Specification for lightweight aggregates for structural concrete, (2009).
30. Kavas T, Christogerou A, Pontikes Y, Angelopoulos G N, Valorisation of different types of boron-containing wastes for the production of lightweight aggregates. Journal of Hazardous Materials. (2011). 185, 1381–1389. https://doi.org/10.1016/j.jhazmat.2010.10.059