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# The effect of additives on the properties of lightweight aggregates produced from clay

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## Abstract

In an attempt to improve the properties, lightweight aggregates were produced from clay with the addition of  $\text{Na}_2\text{CO}_3$ ,  $\text{SiO}_2$ ,  $\text{Fe}_2\text{O}_3$ , and Fe in quantities between 2 and 10 wt% and examined with respect to strength, density and expansion behaviour. The additives were mixed into dry clay powder, water was added and pellets were formed by hand and fired at 1120°C in a chamber furnace. Particle densities of the products ranged from 0.31-0.57 g/cm<sup>3</sup>, porosities from 78-89% and the solid strength from 0,54-1,58 MPa. The addition of  $\text{Na}_2\text{CO}_3$  proved to decrease the viscosity of the glass phase at the surface of the pellets but resulted in a

reduced expansion, irregular shape and pellets sticking together.  $\text{SiO}_2$  addition did not give any major change in properties. The addition of  $\text{Fe}_2\text{O}_3$  increased the pore size in the centre of the pellets, however with insignificant change in strength and density. Adding 5 wt% metallic iron powder led to LWA pellets with increased porosity, reduced density, larger pores and low mechanical strength and could be an useful additive in applications where low density is more important than strength.

*Keywords:  $\text{Na}_2\text{CO}_3$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{SiO}_2$ , Fe, lightweight aggregate, mechanical strength, density, clay*

## **1 Introduction**

Different types of raw materials of natural and artificial origin are used for the production of lightweight aggregates (LWA) [1-5]. The chemical composition of the raw material determines the bloating (expansion) behaviour and the properties of the product [6]. Riley [7] investigated different compositions of clay and defined regimes in the ternary system  $\text{SiO}_2$ - $\text{Al}_2\text{O}_3$ -flux (Riley diagram) where bloating takes place when heated to temperatures up to  $1300^\circ\text{C}$  and given that a gas forming reaction occurs at the same temperature. During firing gas evolution takes place [8] and the material needs to be in a plastic state in order to trap the gases and subsequently expand [7]. Plasticity is obtained by the formation of a glass phase which becomes less viscous above the softening point and allows expansion. The glass phase accounts for a major part of the resulting matrix phase after cooling to ambient temperature [9, 10] and is consequently considered to play an important role with respect to a number of properties of the final LWA-pellet. Previous studies showed different that additives influence the properties of different types of WLA. For example the addition of amorphous  $\text{SiO}_2$  in

form of cullet powder to sewage sludge ash for the production of LWA was reported to enhance the bloating behaviour of the material [11]. Other studies reported the addition of quartz sand to be beneficial for the production of LWA from smectite-rich claystone-marlstone rocks or the addition of waste glass to lignite coal fly ash leading to LWA with advanced physical and mechanical properties [4, 12]. The addition of CaO (a flux) to water reservoir sediments led to an enhanced expansion and pore growth during firing but resulted in a LWA with decreased strength [13].

In order to modify and preferably enhance the properties of LWA produced from another raw material, namely clay with a given composition within the Riley diagram, different additives were chosen and mixed into the raw material and the resulting LWA characterized with respect to mechanical strength, density, micro structure and appearance. The additives in question were  $\text{Na}_2\text{CO}_3$ ,  $\text{SiO}_2$  (silica fume and quartz particles, respectively),  $\text{Fe}_2\text{O}_3$  and Fe.

## **2 Material and methods**

### *2.1 Raw materials*

The utilized raw material for the LWA production was clay blended with 1 wt% waste motor oil as expansion agent. Homogenization of the clay as well as the mixing of oil into the clay was performed by shaft mixers in an industrial production line at Saint-Gobain Weber in Norway. The chemical and mineralogical composition of the clay was determined by an external research company called “IBU-tech advanced materials AG” using gravimetry, wet chemical quantification methods and X-ray diffraction and are given in Tab. 1. The amount of the different additives as well as quality is given in Tab. 2.

mineralogical composition		chemical composition	
Quartz	17	SiO <sub>2</sub>	59
Plagioclase	19	Al <sub>2</sub> O <sub>3</sub>	18
Orthoclase	6	Fe <sub>2</sub> O <sub>3</sub>	7
Amphibole	4	K <sub>2</sub> O	4
Illite/muscovite	40	MgO	3
Chlorite	10	CaO	2
Fe-Oxihydrate	4	Na <sub>2</sub> O	1
		TiO <sub>2</sub>	1
		LOI <sub>(1000°C)</sub>	5

Table 1: Mineralogical and chemical composition of the as received raw clay (in wt%).

composition	description	purity	amount added [wt% of the dry clay]
Na <sub>2</sub> CO <sub>3</sub>	powder	>98 %	2 and 5
SiO <sub>2</sub>	silica fume (amorphous SiO <sub>2</sub> ) powder	>98 %	10
SiO <sub>2</sub>	quartz particles. d <sub>50</sub> - 6.6 μm	>98 %	5
Fe <sub>2</sub> O <sub>3</sub>	hematite powder	>98 %	5
Fe	metallic iron powder	>98 %	5

Table 2: Quality and amount of the different additives applied in the investigation.

## 2.2 LWA manufacturing

Lightweight aggregates were produced manually in the laboratory. The raw clay, containing 1 wt% motor oil, was dried at 105°C for 16 hours and subsequently milled to a powder with an agate hand mortar. Additives were mixed to the dry clay powder and homogenised with a hand mortar. Approximately 23 wt% water (same as the original water content of the as received raw clay) was subsequently added and pellets of constant size and weight were rolled by hand. The green pellets were dried at 105°C for 16 hours, pre-heated for 2 hours at 250°C and finally fired for 8 minutes at temperatures between 1070°C and 1125°C (depending on

composition) in a chamber furnace prior to cooling (approx. 160°C/min) in air to ambient temperature.

### 2.3 Material testing

The average dry particle density,  $\rho_{particle}$ , of each sample series was determined by sand pycnometry. A couple of individual pellets were put into a flask and covered with a known amount of fine sand to measure the volume. The particle density was calculated by dividing the mass of the material by the measured volume.

Helium pycnometry was used to determine the density of the matrix phase,  $\rho_{matrix}$ . Each density measurement was performed by milling a couple of pellets to a particle size  $< 36 \mu\text{m}$  and subsequently assessing the density in an AccuPyc 1330 helium pycnometer from micrometics. The average porosity,  $P$  (in per cent), was calculated for each sample series according to Eq. 1.

$$P = 100 \cdot \left( 1 - \frac{\rho_{particle}}{\rho_{matrix}} \right) \quad (1)$$

The strength of each single pellet was determined by uniaxial compression between 2 parallel rigid platens. The pellet diameter was measured with a calliper before the pellet was placed on the bottom plate of a press. Compression was performed with a constant speed of displacement of 2 mm per minute until a crack ruptured the pellet into at least two pieces. The applied load at failure,  $F_{crit}$ , of several pellets was recorded for each sample-series. The test equipment was a TIRA Test 2420 press including a load cell with a maximum capacity of 1 kN. The platen material was stainless steel. The solid strength,  $\sigma_{crit}$ , of each sample was

calculated from the average load at failure,  $F_{crit}$ , the volume fraction of solid material within the pellet,  $\rho_{particle}/\rho_{matrix}$ , and the average diameter,  $D$ , using Eq. 2.

$$\sigma_{crit} = \frac{F_{crit}}{D^2 \cdot \left(\rho_{particle} / \rho_{matrix}\right)^{2/3}} \quad (2)$$

The result of Eq. 2 is a strength value which is independent of the total porosity,  $P$ , and consequently allows us to assess the strength of the matrix phase as such and compare different samples. The concept of solid strength is further rationalized in [14].

Solid strength results are presented in dependency of the “solid diameter”,  $d$ , which is the theoretical diameter of a pellet with zero porosity and is calculated by Eq. 3.

$$d = D \cdot \left(\frac{\rho_{particle}}{\rho_{matrix}}\right)^{1/3} \quad (3)$$

Prior to catastrophic failure, all pellets will suffer a certain “crumbling” (apparent plastic deformation) at the contact point between platens and pellet. The radius,  $a_c$ , of this “crumbled area” at the point of failure is important to take into account when comparing strength between different samples.  $a_c$  can be approximated from the average anvil deflection at failure,  $x$ , and the initial average radius of a sample,  $R$ , by Eq. 4 assuming a perfect sphere and equal damage/crumbling at the “north- and south-pole” of the pellet.  $a_c$  values between 0.2 and 0.6 are considered to be within an acceptable range [14].

$$a_c = \left(R^2 - \left(R - \frac{x}{2}\right)^2\right)^{\frac{1}{2}} \quad (4)$$

The microstructure of selected samples was investigated using a scanning electron microscope (SEM) from Hitachi (C-3400N). For this type of investigation single pellets were cut in half, embedded in epoxy resin, polished and sputtered with carbon.

### **3 Theory**

#### *3.1 Addition of $\text{Na}_2\text{CO}_3$*

$\text{Na}_2\text{CO}_3$  was used as an additive to potentially reduce the softening temperature of the glass phase that is formed during production.  $\text{Na}_2\text{O}$  is a commonly used flux in the production of soda-lime glasses due to its ability to reduce both the glass transition temperature,  $T_g$ , and the softening temperature,  $T_s$  [15]. Reducing these temperatures for the glass phase in question potentially promotes expansion at even lower temperatures saving both energy and reduce the cost.  $\text{Na}_2\text{CO}_3$  melts at  $850^\circ\text{C}$  and decomposes to  $\text{Na}_2\text{O}$  and  $\text{CO}_2$  at about the same temperature [16]. The kinetics of the decomposition is somehow slow and continues until  $1200^\circ\text{C}$  when heated at a rate of  $10^\circ\text{C}/\text{min}$  [16] and could consequently also contribute to the expansion of LWA.

#### *3.2 Addition of $\text{SiO}_2$*

In contrast to the addition of a flux, an increased  $\text{SiO}_2$  content might lead to an increased viscosity of the glass phase and possibly to the formation of smaller pores and potentially a



more homogeneous pore-distribution within the volume of a pellet.  $\text{SiO}_2$  generally acts as a network former in glasses and thus increases the viscosity and  $T_g$  [17]. It has been reported that LWA incorporating numerous small pores show enhanced mechanical strength compared to LWA with large, irregular pores [8, 18]. However, these alleged, enhanced mechanical properties may be questioned, since the reported strength values are influenced by a variety of parameters (pellet size, total porosity, composition etc.) which severely complicates the assessment of strength variation due to pore distribution alone. The addition of  $\text{SiO}_2$  in form of quartz sand and cullet powder (amorphous  $\text{SiO}_2$ ) was reported to have positive effects on firing temperature, expansion and mechanical properties of different types of LWA [4, 11]. In the present study amorphous  $\text{SiO}_2$  in the form of silica fume and crystalline  $\text{SiO}_2$  in the form of quartz powder were used as additives. Non dissolved quartz particles may also increase the fracture toughness of the glass phase due to crack deflection [19, 20] and in that sense the addition of crystalline particles (quartz) may enhance the mechanical properties of the matrix phase in LWA.

### *3.3 Addition of iron*

Iron is an important component in LWA since  $\text{Fe}_2\text{O}_3$  is one of the main components leading to gas formation at elevated temperatures [8, 21]. Additionally,  $T_g$  and the viscosity of the glass phase are significantly influenced by the oxidation state of iron as observed for iron containing silicate melts where an increased amount of  $\text{Fe}^{3+}$  leads to both a higher viscosity and a higher glass transition temperature [22, 23]. Investigations on the softening temperatures of the matrix phase of clay based LWA showed that, depending on the oxidation state of iron, the softening temperatures varied from about 700°C for a reduced matrix phase (basically  $\text{Fe}^{2+}$ ) to about 950°C (basically  $\text{Fe}^{3+}$ ) for an oxidized matrix phase [24]. Consequently, depending on the oxidation state of the iron-additive the viscosity and softening temperatures of the glass phase might be increased or decreased and expansion

could be promoted due to the presence of additional gas forming components (iron oxide).  $\text{Fe}_2\text{O}_3$  as well as metallic iron were used as additives in order to investigate the influence of additional iron and its oxidation state on the properties of LWA

## 4 Results

### 4.1 Addition of $\text{Na}_2\text{CO}_3$

LWA containing 2 wt%  $\text{Na}_2\text{CO}_3$  fired at  $1125^\circ\text{C}$  are shown in Fig. 1a). Some LWA pellets disintegrated (nearly exploded) during firing or “glued” together due to the formation of a low-viscous phase (melt) at the surface, however all pellets showed a general low degree of expansion. The result of adding 5 wt% is given in Fig. 1b). Some expansion took place but most of the pellets disintegrated into two or more pieces and showed a high degree of open porosity and an irregular particle shape. At even lower firing temperatures (between  $1120^\circ\text{C}$  -  $1070^\circ\text{C}$ ) and an additive content of 5 %, disintegration and sticking could be avoided, the expansion was however poor as evident in Fig. 1c) showing a pellet fired at  $1070^\circ\text{C}$  next to a non fired (“green”) pellet.

The production of LWA from the given raw material, including the addition of  $\text{Na}_2\text{CO}_3$ , did not lead to a satisfactory result and the pellets were not further investigated.

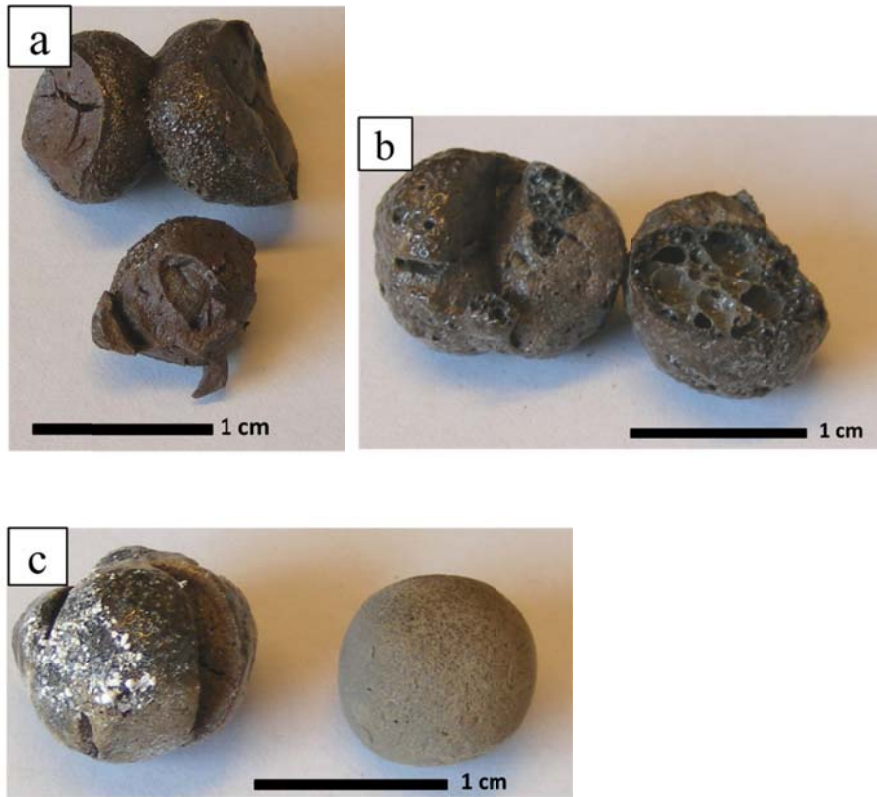


Figure 1: LWA containing  $\text{Na}_2\text{CO}_3$ : a): 2 wt%  $\text{Na}_2\text{CO}_3$ /1125°C burning temperature; b): 5 wt%  $\text{Na}_2\text{CO}_3$ /1120°C burning temperature; c): 5 wt%  $\text{Na}_2\text{CO}_3$ /1070°C burning temperature. The pellet to the right is a non fired (“green”) pellet.

#### 4.2 Addition of $\text{SiO}_2$ , $\text{Fe}_2\text{O}_3$ and Fe

An overview of the main properties of LWA with additional  $\text{SiO}_2$ ,  $\text{Fe}_2\text{O}_3$  and Fe is given in Tab. 3. The standard deviation of the diameter,  $D$ , and the matrix density (approximated to  $0.06 \text{ g/cm}^3$  from the reference sample) are given in brackets. Standard deviations of  $\sigma_{crit}$ ,  $d$ ,  $P$  and  $\rho_{particle}$  are not given since the values result from calculations including the average of several pellets within a sample series and were not measured for every single pellet.

Generally, the standard deviation of all properties is substantial due to the heterogeneous nature of the material.

The main properties ( $\sigma_{crit}$ ,  $P$  and  $\rho_{particle}$ ) are given in Figs. 2-4. The solid strength of LWA does not change noteworthy when adding  $\text{SiO}_2$ ,  $\text{Fe}_2\text{O}_3$  or quartz particles as evident from Fig. 2. However, the addition of metallic iron resulted in a significantly lower strength combined with a distinct increase in porosity (cf. Fig. 3). The high porosity of the 5 % Fe sample means inevitably a low density (Fig. 4) and certainly indicates a high degree of expansion. The 5 % quartz sample showed a slightly lower porosity and higher density than the reference whereas the samples 5 %  $\text{Fe}_2\text{O}_3$  and 10 %  $\text{SiO}_2$  only showed insignificant variances in properties compared to the reference.

sample	diameter $D$ /[mm]	solid diameter $d$ /[mm]	porosity $P$ /[%]	particle density $\rho_{particle}$ / [g/cm <sup>3</sup> ]	matrix density $\rho_{matrix}$ / [g/cm <sup>3</sup> ]	solid strength $\sigma_{crit}$ [MPa]	tested samples	$a_c/R$
reference	16.0 ( $\pm 0.6$ )	8.9	83	0.46	2.73 ( $\pm 0.06$ )	1.48	70	0.24
10% $\text{SiO}_2$	15.8 ( $\pm 0.5$ )	8.8	83	0.44	2.59 ( $\pm 0.06$ )	1.31	25	0.24
5% quartz	15.2 ( $\pm 0.6$ )	9.1	78	0.57	2.62 ( $\pm 0.06$ )	1.58	7	0.21
5% $\text{Fe}_2\text{O}_3$	15.8 ( $\pm 0.6$ )	8.9	82	0.50	2.72 ( $\pm 0.06$ )	1.27	9	0.23
5% Fe	18.6 ( $\pm 0.7$ )	9.0	89	0.31	2.75 ( $\pm 0.06$ )	0.54	20	0.17

Table 3: Overview of the properties of LWA including the different additives.

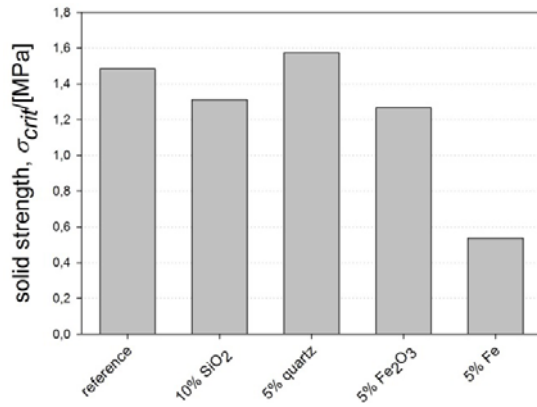


Figure 2: Solid strength of LWA produced with different additives

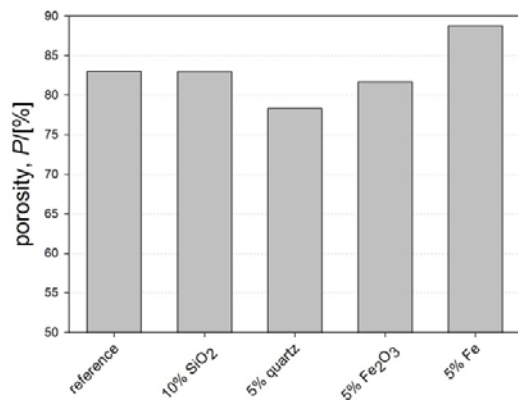


Figure 3: Porosity of LWA produced with different additives

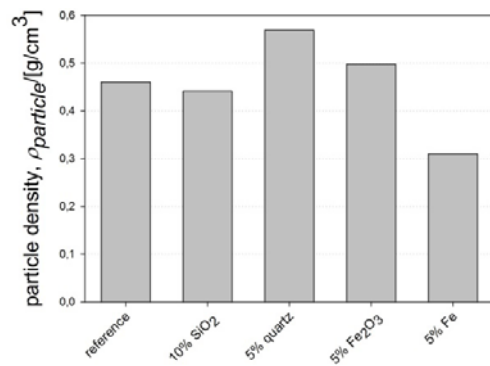


Figure 4: Particle density of LWA produced with different additives

#### 4.3 Addition of $\text{SiO}_2$ (silica fume/quartz) – sample appearance

Fig. 5 shows images from a light microscope for the reference-, 10 %  $\text{SiO}_2$ - and 5 % quartz sample. The appearances of the cross-sections of the pellets are rather similar in coloration, shell thickness, pore size and pore distribution.

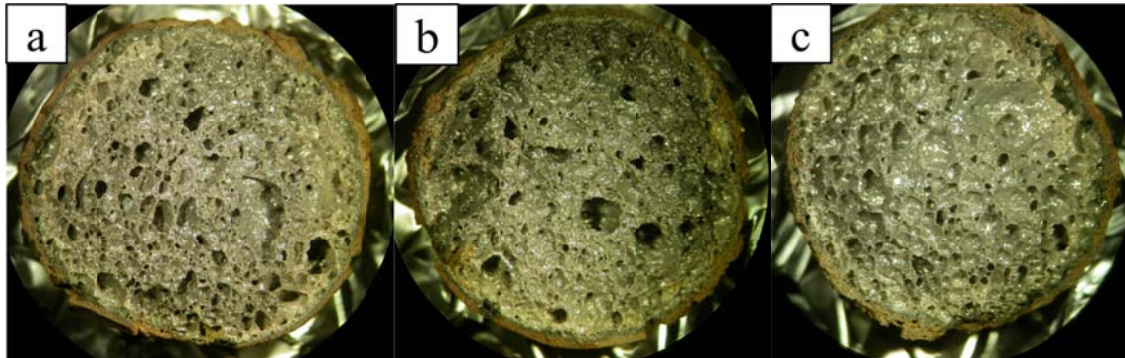


Figure 5: Images from a light microscope of pellets cut in half. a): reference, b): 10 %  $\text{SiO}_2$ , c): 5 % quartz. The diameter,  $D$ , of each pellet is 15.5 mm.

#### 4.4 Addition of iron ( $\text{Fe}_2\text{O}_3/\text{Fe}$ ) – sample appearance

Light microscope images of the 5 %  $\text{Fe}_2\text{O}_3$ - and 5 % Fe sample are given in Fig. 6. The addition of 5 %  $\text{Fe}_2\text{O}_3$  leads to larger pores in the centre of the pellet compared to the reference whereas the total porosity,  $P$ , and diameter,  $D$ , did not change significantly (cf. Fig. 3 and Tab. 3). The colour of the pellet changes in the following way: The shell has a red appearance and is positioned next to a black “transition zone” located between the shell and the grey-green- appearing core. The pore size increases from the almost dense shell (low porosity) via the transition zone (small pores) to the core (large pores).

The addition of metallic iron led to larger pores throughout the whole pellet compared to the reference. The shell is very thin and the samples expanded more than the reference (89 % total porosity) also resulting in a larger diameter,  $D$  (cf. Tab. 3). The colour is grey-green in the centre and a thin black “transition zone” is visible between the core and the red-brown shell as observed in the sample with 5 %  $\text{Fe}_2\text{O}_3$ . However, the shell and the transition zone are much thinner in the 5 % Fe sample than in the 5 %  $\text{Fe}_2\text{O}_3$  sample.

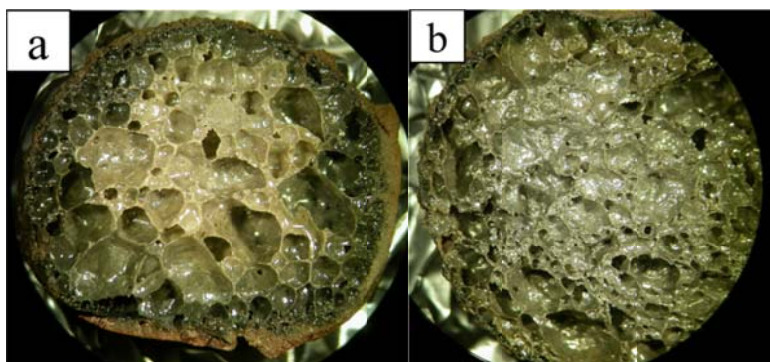


Figure 6: Images from a light microscope of pellets cut in half with the addition of 5 %  $\text{Fe}_2\text{O}_3$  (a) and 5 % Fe (b). The cross section of a) is 15.5 cm and 16.5 cm for b).

#### 4.5 Addition of iron ( $\text{Fe}_2\text{O}_3/\text{Fe}$ ) – investigation of the microstructure by SEM

Backscattered electron images of the shell and core of the samples 5 %  $\text{Fe}_2\text{O}_3$  and 5 % Fe are given in Fig. 7. The matrix-phase of the core looks very similar for both the addition of  $\text{Fe}_2\text{O}_3$  and Fe (cf. Fig 7a) and 7c)). The glass phase (continuous grey phase) includes several almost spherical pores and different types and shapes of crystalline inclusions. Smaller, bright white spots are present in the 5 % Fe sample (Fig. 7a)) indicating the presence of metallic iron. A

significant difference between the two iron containing samples is observed in the shell: The 5 % Fe sample incorporates small pores which are surrounded by iron rich areas indicating that the iron caused expansion. The 5 %  $\text{Fe}_2\text{O}_3$  sample also shows spots of higher atomic number (indicated by bright colours in Fig. 7 d)) in the shell but no expansion (pores) was observed in these areas.

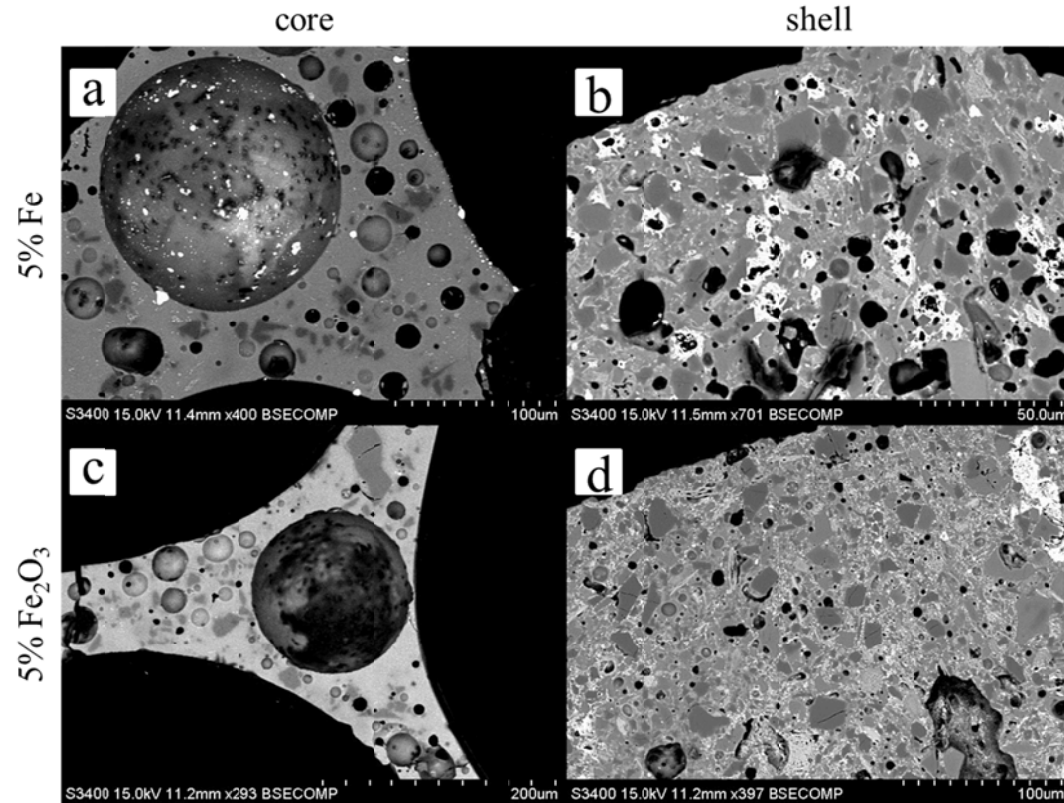


Figure 7: BSE images of the shell and core of the samples 5%  $\text{Fe}_2\text{O}_3$  and 5% Fe.

## 5 Discussion

### 5.1 Reliability of data

In order to generally obtain reliable and comparable strength values from the single pellet compression test the ratio of  $a_c/R$  (radius of the crushed area on top and bottom of the sample



divided by radius of the sample) should be between 0.2 and 0.6 [14, 25, 26]. This requirement is fulfilled for all samples.

### *5.2 Addition of $\text{Na}_2\text{CO}_3$*

The viscosity of the glass phase seems to be reduced through the addition of  $\text{Na}_2\text{CO}_3$  resulting in a glassy appearing shell. This may be positive since a molten surface would lead to a LWA with low permeability (water absorption) and potentially enhanced strength. However, the same effect also causes the pellets to stick together which would make the production of this type of LWA on an industrial scale rather demanding. Additionally,  $\text{Na}_2\text{CO}_3$  containing pellets tended to explode during firing which is most likely caused by the additional gas formation due to the decomposition of  $\text{Na}_2\text{CO}_3$  resulting in irregular shaped pellets, often showing a high degree of open porosity and consequently leading to a product with less favourable properties such as high water absorption and high bulk density.

### *5.3 Addition of $\text{SiO}_2$*

The addition of  $\text{SiO}_2$  in the form of silica fume and quartz particles did not lead to significant differences in appearance or properties of the resulting LWA. The raw clay contains already a high amount of  $\text{SiO}_2$  and a moderate increase seems not to affect the properties of the resulting LWA notably, as supported by a pore structure and strength basically independent of the enhanced silica content (Figs. 2-5). Furthermore, adding 5% quartz does not seem to affect the strength (Fig. 3) although there is a tendency of less porosity (Fig. 4) and increased density (Fig. 5). Based on the small number of tested samples, it is not possible to draw any

unambiguous conclusions, however, it seems that there is no strong correlation between strength and silica content in the composition interval investigated.

#### *5.4 Addition of iron ( $Fe_2O_3 / Fe$ )*

The addition of  $Fe_2O_3$  did not give any noteworthy change in density, pellet size, porosity or strength. However, the pore structure changed markedly to larger pores in the centre of the pellet compared to the reference. The colour of the matrix phase also changed due to changes in the oxidation state of iron [8], whereas a red colour indicates a high amount of  $Fe^{3+}$ , a grey-green colouration points to a high amount of  $Fe^{2+}$  and black an intermediate oxidation state. The gradient in oxidation states is a consequence of a highly reducing atmosphere (basically CO) inside the pellet, caused by the carbothermal reduction of  $Fe_2O_3$ , and an increasingly more oxidizing atmosphere towards the surface of the pellet due to oxygen diffusing into the pellet as a consequence of firing in air. The resulting enhanced content of iron in a low oxidation state in the centre of the pellet leads to a lower viscosity of the glass phase compared to the reference as indicated by [22-24] causing the formation of large pores (cf. Fig 6a)). Additionally, the added  $Fe_2O_3$  dissolved into the glass phase as evident in Fig. 7c) where no iron rich areas were observed. In contrast,  $Fe_2O_3$  was not completely dissolved in the glass phase in the shell leaving some iron rich spots in the matrix phase (cf. Fig. 7d)).

The addition of metallic iron affected almost all properties compared with the reference. The high porosity and expansion are the result of a low-viscosity glass phase caused by the low oxidation state of iron. Accordingly, the conditions in the core are quite similar to what has been described for the addition of  $Fe_2O_3$ . However, the shell of the 5% Fe sample becomes

thinner compared both to the reference and the 5% Fe<sub>2</sub>O<sub>3</sub> sample and expansion takes place even in the shell itself as shown in Fig. 7b). It is likely that metallic iron is oxidized during firing and subsequently reduced again leading to gas formation and expansion in the shell. The general lower oxidation state of iron throughout the whole pellet leads to a lower viscosity of the glass phase and enables an overall higher degree of expansion resulting in a high porosity. Nevertheless, it should be noted that the molar fraction of Fe is about 30 % higher in the 5 % Fe sample than in the 5 % Fe<sub>2</sub>O<sub>3</sub> sample which may affect some of the observed variations between these two samples. The pellets of the 5 % Fe sample become very fragile and show a poor mechanical strength due to the high porosity and the thin shell, consistent with observations previously reported by Bernhardt et al. [14].

#### 4 Conclusions

Different additives have been tested aiming at improving both the production process and the properties of lightweight expanded clay aggregates. Na<sub>2</sub>CO<sub>3</sub> proved to decrease the viscosity of the glass phase at the surface of the pellets but resulted in low expansion, irregular shape and pellets sticking together. Consequently, the production of good quality LWA was not possible with the addition of Na<sub>2</sub>CO<sub>3</sub> and the initial goal of reducing the production temperature was not achieved.

The addition of SiO<sub>2</sub>, whether amorphous or crystalline, affected the properties in question insignificantly in the composition interval investigated.

Iron was found to influence the properties of LWA significantly. Fe<sub>2</sub>O<sub>3</sub> addition resulted in larger pores in the centre part of the pellet but did not affect the strength, density or total

porosity. On the other hand, the addition of metallic iron powder gave a major increase in expansion and accordingly reduced both particle density and mechanical strength. Fe powder could, potentially, be used as an effective additive to decrease the density of LWA in applications where low density is more important than strength. Additionally, the modification of the porosity of LWA might be an interesting tool to influence the application of LWA for internal curing of concrete.

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### **References**

- [1] González-Corrochano B, Alonso-Azcárate J, Rodas M. Characterization of lightweight aggregates manufactured from washing aggregate sludge and fly ash. *Resources, Conservation and Recycling*. 2009;53(10):571-81.
- [2] Cheeseman CR, Makinde A, Bethanis S. Properties of lightweight aggregate produced by rapid sintering of incinerator bottom ash. *Resources, Conservation and Recycling*. 2005;43(2):147-62.
- [3] de' Gennaro R, Cappelletti P, Cerri G, de' Gennaro M, Dondi M, Langella A. Zeolitic tuffs as raw materials for lightweight aggregates. *Applied Clay Science*. 2004;25(1-2):71-81.
- [4] Fakhfakh E, Hajjaji W, Medhioub M, Rocha F, López-Galindo A, Setti M, et al. Effects of sand addition on production of lightweight aggregates from Tunisian smectite-rich clayey rocks. *Applied Clay Science*. 2007;35(3-4):228-37.
- [5] Bernhardt M, Tellesbø H, Justnes H, Wiik K. Fibre reinforced lightweight aggregates. *Journal of the European Ceramic Society*. submitted May 2013.
- [6] Bajare D, Korjakins A, Kazjonovs J, Rozenstrauha I. Pore structure of lightweight clay aggregate incorporate with non-metallic products coming from aluminium scrap recycling industry. *Journal of the European Ceramic Society*. 2012;32(1):141-8.
- [7] Riley CM. Relation of chemical properties to the bloating of clays *Journal of the American Ceramic Society*. 1951;34(4):121-8.

- [8] Decler J, Viaene W. Rupelian Boom clay as raw material for expanded clay manufacturing. *Applied Clay Science*. 1993;8(2-3):111-28.
- [9] de Gennaro R, Cappelletti P, Cerri G, de' Gennaro M, Dondi M, Graziano SF, et al. Campanian Ignimbrite as raw material for lightweight aggregates. *Applied Clay Science*. 2007;37(1-2):115-26.
- [10] Qi Y, Yue Q, Han S, Yue M, Gao B, Yu H, et al. Preparation and mechanism of ultra-lightweight ceramics produced from sewage sludge. *Journal of Hazardous Materials*. 2010;176(1-3):76-84.
- [11] Tsai CC, Wang KS, Chiou JJ. Effect of SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>-flux ratio change on the bloating characteristics of lightweight aggregate material produced from recycled sewage sludge. *Journal of Hazardous Materials*. 2006;134(1-3):87-93.
- [12] Kourti, I. and C.R. Cheeseman, Properties and microstructure of lightweight aggregate produced from lignite coal fly ash and recycled glass. *Resources, Conservation and Recycling*, 2010. 54(11): p. 769-775.
- [13] Liao, Y.C. and C.Y. Huang, Effects of CaO addition on lightweight aggregates produced from water reservoir sediment. *Construction and Building Materials*, 2011. 25(6): p. 2997-3002.
- [14] Bernhardt M, Tellesbø H, Justnes H, Wiik K. Mechanical properties of lightweight aggregates. *Journal of the European Ceramic Society*. 2013, vol. 33: p. 2731-43.
- [15] Shelby JE, Mesko MG, Shulman H, Edson DL. Properties of soda-lime-silica glasses. 2004. p. 371-8.
- [16] Kim JW, Lee HG. Thermal and carbothermic decomposition of Na<sub>2</sub>CO<sub>3</sub> and Li<sub>2</sub>CO<sub>3</sub>. *Metallurgical and Materials Transactions B: Process Metallurgy and Materials Processing Science*. 2001;32(1):17-24.
- [17] Shelby JE. *Introduction to glass science and technology*. Cambridge: Royal Society of Chemistry; 2005.
- [18] Wang X, Jin Y, Wang Z, Nie Y, Huang Q, Wang Q. Development of lightweight aggregate from dry sewage sludge and coal ash. *Waste Management*. 2009;29(4):1330-5.
- [19] Richerson DW. *Modern Ceramic Engineering* third ed: Taylor and Francis group; 2006.
- [20] Rawlings RD. Glass-ceramic matrix composites. *Composites*. 1994;25(5):372-9.
- [21] Kolesnikov EA. Bloating of fusible clays. *Glass and Ceramics*. 1975;31(5):354-6.
- [22] Dingwell DB. Redox viscometry of some Fe-bearing silicate melts. *American Mineralogist*. 1991;76(9-10):1560-2.
- [23] Liebske C, Behrens H, Holtz F, Lange RA. The influence of pressure and composition on the viscosity of andesitic melts. *Geochimica et Cosmochimica Acta*. 2003;67(3):473-85.
- [24] Bernhardt M, Tellesbø H, Justnes H, Wiik K. The effect of heat treatment and cooling rate on the properties of lightweight aggregates. *Journal of the European Ceramic Society*. 2014, vol. 34, Issue 5, p. 1353-1363.
- [25] Shipway PH, Hutchings IM. Attrition of brittle spheres by fracture under compression and impact loading. *Powder Technology*. 1993;76(1):23-30.
- [26] Shipway PH, Hutchings IM. Fracture of brittle spheres under compression and impact loading. *Philosophical Magazine A*. 1993;67(6):1389-404.