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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 Na_2CO_3, SiO_2, Fe_2O_3, and Fe in quantities between 2 and 10 wt% and examined with respect to strength, density and expansion behavior. 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 to 0.57 g/cm^3, porosities from 78% to 89% and the solid strength from 0.54 to 1.58 MPa. The addition of Na_2CO_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. SiO_2 addition did not give any major change in properties. The addition of Fe_2O_3 increased the pore size in the center 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 a useful additive in applications where low density is more important than strength.

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) behavior and the properties of the product [6]. Riley [7] investigated different compositions of clay and defined regimes in the ternary system SiO_2–Al_2O_3–flux (Riley diagram) where bloating takes place when heated to temperatures up to 1300 °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 that different additives influence the properties of different types of LWA. For example, the addition of amorphous SiO_2 in form of cullet powder to sewage sludge ash for the production of LWA was reported to enhance the bloating behavior 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 also 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, microstructure and appearance. The additives in question were Na_2CO_3, SiO_2 (silica fume and quartz particles, respectively), Fe_2O_3 and metallic Fe.

2. Materials and methods

2.1. Raw materials

The utilized raw material for the LWA production was clay blended with 1 wt% waste motor oil as an expansion agent.

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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 Table 1. The amount of the different additives as well as their quality is given in Table 2.

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 h and subsequently milled to a powder by hand with an agate mortar. Additives were mixed into the dry clay powder and homogenized 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 h, pre-heated for 2 h at 250 °C and finally fired for 8 min 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 (pellets) by their measured volume.

Helium pycnometry was used to determine the density of the matrix phase, ρ_{matrix}. Each density measurement was performed by milling a couple of pellets to a particle size <36 μm 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).

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

The strength of each single pellet was determined by uniaxial compression between 2 parallel rigid platens. The pellet diameter was measured with a caliper before the pellet was placed on the bottom plate of a press. Compression was performed at a constant rate of displacement of 2 mm/min 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 TIRATest 2420 press including a load cell with a maximum capacity of 1 kN. The platen material was stainless steel. The solid strength, σ_{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 pellet's average diameter, D, using Eq. (2).

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

The result of Eq. (2) is a strength value which is independent of the total porosity, P, and consequently allows one 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).

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

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].

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

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

3. Theory

3.1. Addition of Na_2CO_3

Na_2CO_3 was used as an additive to potentially reduce the softening temperature of the glass phase that is formed during production. Na_2O 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 energy and reducing the cost. Na_2CO_3 melts at 850 °C and decomposes to Na_2O and CO_2 at about the same temperature [16]. The kinetics of the decomposition is somehow slow and continues until 1200 °C when heated at a rate of 10 °C/min [16] and could consequently also contribute to the expansion of LWA.

3.2. Addition of SiO_2

In contrast to the addition of a flux, an increased 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. 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

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

Mineralogical composition		Chemical composition	
Quartz	17	SiO_2	59
Plagioclase	19	Al_2O_3	18
Orthoclase	6	Fe_2O_3	7
Amphibole	4	K_2O	4
Illite/muscovite	40	MgO	3
Chlorite	10	CaO	2
Fe-Oxihydrate	4	Na_2O	1
		TiO_2	1
		$LOI_{(1000°C)}$	5