











Unraveling the expansion mechanism in lightweight aggregates: Demonstrating that bloating barely requires gas

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Abstract

Lightweight aggregate bloating process has been studied by a simple experiment using an archetypal clay to know the actual amount of gas involved in expansion. Considering the relationship between gas loss (LOI) and volumetric changes over time, three main stages are identified: 1) a preheating stage of massive gas loss (close to 80% of the total) with hardly any volumetric change; 2) a very brief transition stage, in which sintering (shrinkage) and closed (micro)porosity formation begin, accompanied by a sudden gas loss (close to 100% of the total); 3) the bloating stage itself, in which an appropriate viscosity is reached, allowing the available residual gas (<0.1 wt%) to increase the aggregate volume due to the growth in size of the micropores formed in the transition phase and probably also to the development of new porosity. Therefore, the proportion of gas-generating components estimated to obtain a highly expanded lightweight aggregate would be much lower than previously thought: e.g., only 0.06 to 0.2% of carbonates (calcite or dolomite) or 0.2 to 1% of Fe₂O₃ would actually be involved in bloating. These results suggest that obtaining an adequate viscosity appears to be much more decisive for bloating than gas release capacity.

Introduction

As defined in the European standards [1], the term *aggregate* refers to a granular material primarily intended for construction. The aggregates industry is the largest provider of raw materials for building construction and infrastructures, being also fundamental in other industrial sectors as well as in the protection of the

environment. All this entails that aggregate is the second most consumed raw material, surpassed only by water, which makes the aggregate sector highly strategic [2].

Among the different types of aggregates, *lightweight aggregate* (LWA) is characterized by its low density, exhibiting values of less than 1.20g/cm^3 or 2.00g/cm^3 of loose bulk density or particle density (oven dry density), respectively [1].

Four main classes of lightweight aggregates can be defined as a function of the origin and type of processing: *recycled, industry by-products, natural* and *artificial* [1], [3], [4], [5]. The 'artificial' type is the most widespread, representative and growing category, considering that most of the LWA industry as well as the associated research are focused on this line. Artificial lightweight aggregate is characterized by its mineral and inert nature, achieved as a result of a man-made transformation process. Therefore, both artificially manufactured lightweight aggregates from natural resources and those produced from residues and industrial by-products are included in this category. As for the former, the most commonly synthesized LWAs from natural raw materials are expanded clay, shale and slate [3], [4]. Concerning the latter, there is an extensive list of both organic and inorganic wastes that can be transformed into lightweight aggregates, something that is reflected in the review of Dondi et al. [4] on this topic.

Artificial lightweight aggregates can be obtained according to four different manufacturing protocols: *autoclaving, cold bonding, geopolymerization* and *sintering* [3]. Sintering (also called *firing*) is the most common of the four processes mentioned above. In general, the two stages governing such a manufacturing protocol are the granulation or pelletizing and, of course, the heat treatment. [6]. As far as granulation is concerned, the raw materials are ground to a very fine particle size and then mixed in certain proportions if more than one type of raw material is used. The resulting material is then blended with an appropriate proportion of water, which must provide the adequate consistency for the subsequent pelletizing step, either by extrusion or agglomeration. On an industrial level, agglomeration is carried out by different mechanisms, such as rotary discs, pans or drums, cones or mixer-type pelletizers [7]. After the granulation process, the 'green' pellets are subjected to the heat treatment, first for drying and then for sintering, which is the most critical stage of the manufacturing process.

In contrast to the gentle heating ramp used in firing traditional ceramics, such as brick or tiles, the studies published to date consider that the development of a porous structure in LWAs usually requires rapid heating to high temperatures by thermal shock. Thus, on the one hand, the surface of the granulate vitrifies rapidly, hindering the escape of the evolved gases to the outside [8], and, on the other hand, the mineral phases have time to become sufficiently viscous before all the gas-generating components are decomposed, so that the gases formed later can be trapped in this viscous mineral matrix [6], [8]. Typical firing temperatures in lightweight aggregates range from 1050°C to 1250°C , while the dwell time in the kiln is usually 3 to 20min [6]. Obviously, both the temperature applied and the residence time in the kiln will vary depending on the particular characteristics of the raw material and the intended properties of the sintered aggregate.

Regarding the available technologies for LWA sintering, at present, some systems such as the fluidized bed or the travelling grate are almost in disuse. There are some exceptions, such as Lytag® lightweight aggregate, which is manufactured from coal fly ash using a sinter strand, being one of the most successful commercial products within the LWA industry. However, the most widely used lightweight aggregate manufacturing system by manufacturers relies on firing in rotary kilns [9], which has undergone almost no change since it

was defined several decades ago [8], [9]. LWA production in tubular rotary kilns consists of two main stages [8]: first, the granules are preheated at 200–800 °C in the first kiln tube stretch. Then, due to the rotation and inclination of the kiln tube, the granules are gradually passed to the middle section of the tube, where the maximum temperature is reached, at which the material expands. There are two variants of the rotary kiln firing process [8]: *dry* manufacturing or *wet* manufacturing. Dry production means that the pellets are placed in the kiln without moisture, for which they have been previously dried. The wet protocol, by contrast, involves placing the pellet in the kiln directly containing the extrusion or granulation water, which is gradually lost in the first tube section during preheating.

Therefore, the manufacturing procedure described above is based on the self-bloating capacity of the material when subjected to high temperatures. The currently accepted theory explaining such an expansion mechanism is more than sixty years old [10], [11]. This is based on the fact that the porous and light structure of LWAs develops if two conditions occur simultaneously during firing [10], [11].

- i) The formation of a viscous mineral matrix, but without reaching total melting conditions.
- ii) The release of gases (e.g. CO₂, CO, H₂O, H₂, O₂, SO₂ or Cl₂) from the alteration and thermal decomposition of organic phases (e.g. organic matter) and/or mineral species (e.g. carbonates, sulfides, phyllosilicates, chlorides or ferrous minerals, among others [8], [10], [11]).

Therefore, only if the two conditions described above occur at the same time, the gases released during firing can be trapped in the viscous matrix, generating the typical porous structure of lightweight aggregates. This usually leads to a significant increase in size compared to the volume of the original pellet, a phenomenon known as *bloating* or *expansion*.

However, the relationship between the exact amount of gas released at each stage and the variation in size of the aggregate has never been investigated. To this end, a very simple but at the same time very revealing experiment has been carried out. An archetypal clay has been used to produce lightweight aggregates by thermal treatment. The volumetric changes, the total LOI and the LOI suffered during rotary kiln firing have been related to each other in order to understand the different phases occurred during lightweight aggregate formation, with special attention to the amount of gas involved in bloating.

Section snippets

Sampling and initial preparation of the clay

For this study, an Italian red clay (Zocca, Modena province) currently extracted for the ceramic industry (Escavazioni Baroni S.R.L.) has been used. It was selected for presenting suitable characteristics for bloating without the need for any additive according to previous tests. Once in the laboratory, the clay was dried in an oven at 105 °C. Subsequently, it was ground using a Restch® SK 100/C Spezialstahl arm mil, whereby the milled clay had a particle size of less than 200 μm [12]...

Characterization of the clay

The...

Characteristics of the clay

The studied clay presents a very fine particle size (average size of $9.1\ \mu\text{m}$, d_{50} of $4.4\ \mu\text{m}$ and only 1.1% of fraction $>63\ \mu\text{m}$) and a moderate-high plasticity ($LL=44.7$, $PI/LL=0.60$, $T_{\text{max}}=34.4\ \text{kJ/m}^3$) (Table 1). This makes it a very workable material for extrusion and granulation. These results agree with the mineralogy of the sample, rich in phyllosilicates such as illite and kaolinite (Table 2). The iron content is relatively high (7.2%; Table 2), which is generally a key component in the...

Conclusions

The bloating mechanism of lightweight aggregates has been studied in a novel way through a very simple experiment. The main objective was to know the amount of gas and gas-generating components actually involved in lightweight aggregate expansion. The monitoring of the loss on ignition and the volumetric changes suffered by an archetypal clay with respect to the firing time have allowed the following conclusions to be drawn:

The bloating process consists of three main stages:

- A preheating stage...

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CRedit authorship contribution statement

José Manuel Moreno-Maroto: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Supervision, Validation, Visualization, Writing - original draft, Writing - review & editing.

Carlos Javier Cobo-Ceacero: Data curation, Investigation, Writing - review & editing. **Manuel Uceda-Rodríguez:** Data curation, Investigation, Writing - review & editing. **Teresa Cotes-Palomino:** Funding acquisition, Project administration, Resources, Writing - review & editing. **Carmen Martínez García:**...

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper....

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...However, as has been experimentally demonstrated in a previous investigation [19], almost all of the gases generated

actually escape out of the aggregate. In fact, according to the work published by Moreno-Maroto et al. [19], contrary to what was thought until then, the amount of gas directly involved in the expansion process of the lightweight aggregate is negligible, representing less than 0.1 wt. % of the original sample....

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