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Effect of prefiring and firing dwell times on the properties of artificial lightweight aggregates

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HIGHLIGHTS

- Expansion of artificial aggregates largely depends on the prefiring dwell time.
- Shorter prefiring and firing times produce less dense artificial aggregates.
- Prefiring and firing times affect the formation of a vitrified external layer.
- Prefiring and firing times affect the formation of longer internal pores.

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ABSTRACT

The effects of the prefiring and firing dwell times on the chemical, physical and microstructural properties of artificial lightweight aggregates (LWAs) produced from mining and industrial waste with a tubular rotary kiln were determined. It was observed that 1.5 and 3 min of prefiring and firing dwell times, respectively, were more adequate than 5 and 4 min for producing less dense artificial aggregates (0.98 g/cm^3 vs 1.64 g/cm^3). The LWAs manufactured with shorter dwell times could be used in gardening and horticulture, geotechnical applications and/or as insulation materials. LWAs manufactured with longer dwell times could also be used for insulating lightweight concrete and/or prefabricated lightweight structures.

1. Introduction

Coal combustion products are highly important industrial by-products in developed countries and a total of 52 × 10^6 tons were produced in 15 EU states in 2009 [1]. The mining of sulfide ore deposits produces large quantities of hazardous waste due to their heavy metal content and the formation of acid drainage [2]. Therefore, it is very important to develop new technologies that allow this waste to be recycled into added-value products.

According to the UNE-EN-13055-1 standard [3], a *lightweight aggregate* (LWA) is defined as a granular material with a loose bulk density (ρ_b) not exceeding 1.20 g/cm^3 or with a particle density not exceeding 2.00 g/cm^3.

Production of artificial LWAs (formed in a ceramic process in which materials that have the ability to expand are rapidly heated at high temperatures) from waste is deemed to be a very satisfactory environmental alternative to dealing with this waste, since a raw material, with no value, becomes a product with important applications.

Abbreviations: BI, bloating index; *BM*, bulk mineralogy; ρ_b, loose bulk density; ρ_a, apparent particle density; ρ_d, dry particle density; WA_{24h}, water absorption after 24 h; *S*, compressive strength; LWAs, lightweight aggregates; *SQ-63*, clay-silt fraction of a polluted mine soil; *FA*, coal combustion products; *XRD*, X-ray diffraction; *ICP-AES*, inductively coupled plasma-atomic emission spectroscopy; LOI_{rm}, loss on ignition of raw materials; *TC*, total carbon content; *OC*, organic carbon content; *IC*, inorganic carbon content; *Si/ΣF*, SiO_2/ΣFluxing ($CaO + MgO + K_2O + Na_2O + FeO + Fe_2O_3$) ratio; *S75F25*, a 75% (wt) mixture of the <63 μm fraction of the contaminated mine soil and 25% (wt) of the coal combustion products; *LWA-4.5*, LWAs prefired for 1.5 min and fired for 3 min; *LWA-9*, LWAs prefired for 5 min and fired for 4 min; LOI_1, loss on ignition of the granules in zone 1; LOI_2, loss on ignition of the granules in zone 1 and zone 2 together; LOI_{total}, total loss on ignition of the granules; *SEM*, scanning electron microscope; *EDX*, energy-dispersive X-ray; *EMPA*, electron microprobe analyses; *BSEM*, back-scattered electron microscope; *S50F50*, a 50% (wt) mixture of the <63 μm fraction of the contaminated mine soil and 50% (wt) of the coal combustion products; *S25F75*, a 25% (wt) mixture of the <63 μm fraction of the contaminated mine soil and 75% (wt) of the coal combustion products; *TG–DTA*, thermogravimetry–differential thermal analysis.

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Thermal and acoustic insulation, geotechnical applications, gardening, production of precast structural units and structural lightweight concrete are some of the potential applications for lightweight aggregates [4–6] although the applications they can have depend on their chemical and physical properties. Moreover, physical properties, such as particle density, compressive strength (*S*) and water absorption (WA_{24h}), are intrinsically correlated with the LWA microstructures [7–9].

In the ceramic industry, two types of heating techniques usually take place [10]:

(i) One-step heating: prefiring and firing are simultaneous.
(ii) Two-step heating: prefiring precedes firing. The main objective of this thermal pretreatment is to stop the dried ceramic bodies subjected to rapid "flash heat", deteriorating, to prevent them from exploding and forming black core during the final firing process [11].

Prefiring and firing dwell times affect the properties of artificial lightweight aggregates [10]. Therefore, in order to improve the production of lightweight aggregates which show the properties required for specified applications, it is necessary to research the relationship between the LWA properties and the prefiring and firing dwell times when they are being manufactured.

Stamboliev [12] carried out one of the first studies with controlled prefiring of spherical granules at approximately 550–600 °C. Mun [13] manufactured lightweight aggregates in a two-stage heating process obtaining products with a minimum loose bulk density of approximately 0.6 g/cm^3. González-Corrochano et al. [11,14] also used two-stage heating to produce lightweight aggregates, recycling different types of waste. Later, Qi et al. [10] studied the prefiring mechanism, bloating phenomenon and the optimum conditions for producing ultra-lightweight ceramics from sewage sludge. Then, Yue et al. [15] researched the lightweight ceramic sintering mechanism in which there are different firing temperatures and prefiring treatment. With a few exceptions [10], most of the research carried out with the two-step heating process only employed one time and/or one temperature range for the prefiring step [11,13,15], as more importance was given to the second step. In this study, two different prefiring dwell times will be tested.

The aim of this paper was to study the combined effects of prefiring and firing dwell times on the chemical, physical and microstructural properties of artificial lightweight aggregates produced from mining and industrial waste, including polluted mine soil and coal combustion products. Additionally, this waste was recycled in order to obtain useful materials, such as the aforementioned lightweight aggregates.

2. Raw materials

Two raw materials/types of waste were used to produce the LWAs:

– A clay–silt fraction of a polluted mine soil (*SQ-63*). Bulk samples from contaminated soil were taken from the area of the "San Quintín" Pb–Zn mine, located in the Alcudia Valley (Ciudad Real, Spain) with the main ore minerals extracted from this mine being galena (PbS) and sphalerite (ZnS). When the mine closed, Pb and Zn were recovered by floating the low-grade mineral in the tailings. The sludge generated in this process was pumped from the flotation tanks into the waste ponds which produced detrimental effects on the region (large areas were covered with contaminated sediments [16]) and, subsequently, high levels of heavy metals in soils belonging to mining areas and the surrounding arable and grazing lands were detected [16]. After sampling, wet sieving to <63 μm of this mine soil was carried out.
– Coal combustion products (*FA*). The sample was provided by the Aliaga thermal power station (Teruel, Spain), which is no longer in operation. This residue was disposed of in a slagheap with a volume of 11×10^6 tons and the sample was oven dried at 60 °C for several days.

3. Materials and methods

3.1. Characterization of the raw materials

Bulk mineralogy (*BM*) was carried out by X-ray diffraction (*XRD*; [17,18]) after grinding and homogenizing the raw materials to <53 μm. Random-oriented powders were then examined on a *X'Pert Pro* diffractometer, using Cu Kα radiation, a 45 kV accelerating voltage and a 40 mA current. The slit system (soller-mask-divergence-anti-scatter slits) was made up of 0.04 rad–10 mm–1/8°–1/4° slits. The step size was 0.01 (°2θ) and the time per step was 40 s (scan speed of 0.05°2θ).

After fusion with lithium metaborate at 1000 °C and acid dissolution [19], the chemical composition was analyzed using inductively coupled plasma-atomic emission spectroscopy (*ICP-AES*, *Thermo Electron 6500 ICAP*). Loss on ignition (LOI_{rm}) during fusion was then determined as being the percentage weight difference in heated samples at 1100 °C over 24 h.

Then, the total, organic and inorganic carbon contents (*TC*, *OC* and *IC*, respectively) were determined using a *Shimadzu® TOC-VCSH* analyser.

To evaluate the bloating behavior of the raw material and its potential mixtures, its different compositions were plotted on the ternary Riley diagram [20]. Then, the SiO_2/ΣFluxing (CaO + MgO + K_2O + Na_2O + FeO + Fe_2O_3) ratio (*Si*/*ΣF*) was calculated as it showed the ability of the material to form a mass viscous enough to trap released gases [21,22].

3.2. LWA manufacturing

On the basis of the results obtained in the physicochemical characterization of *SQ-63* and *FA*, a 75% (wt) mixture of the <63 μm fraction of the contaminated mine soil and 25% (wt) of the coal combustion products (*S75F25*) was chosen. This mixture has a composition that is expected to be a good material source for LWA production since it is located within the "expansion area" in the Riley diagram [20]. On the contrary and as the results will show, the 50% *SQ-63*, 50% *FA* (*S50F50*) and the 25% *SQ-63* and 75% *FA* (*S27F75*) mixtures show compositions outside this expansion area. The selected mixture was milled to a grain size of less than 200 μm [23] and homogenized using a *Restch® SK 100*/*C Spezialstahl* arm mill.

The plasticity of *S75F25* was then determined by calculating Atterberg's limits, using the procedure established in the Spanish regulations: Norm UNE-103-103 [24] and Norm UNE-103-104 [25].

Water was then added to the ground mixture to a level 2–3% above the plastic limit and then extruded using a *Nannetti®* pneumatic extruder. Next, to produce approximately spherical 8–10 mm diameter granules, cylinders of 1.5 cm of length were cut and rolled by hand.

The green pellets were next dried for 48 h at room temperature and then they were put in an oven at 105 °C for 48 h.

Heating of granules was carried out in a tubular rotary kiln (*TOR 120-14*, *Nannetti®*) with a Al_2O_3 refractory tube, with an internal diameter of 6 cm and length of 120 cm. Three areas could be differentiated in the tube depending on the temperature reached: a central area (*firing area* or *zone 2*), where the temperature was the highest (the temperature the user selected), and two lateral areas, with lower temperatures, one located at the tube entrance (*prefiring area* or *zone 1*) and one situated at the exit (*cooling area* or *zone 3*). Temperature in these lateral areas depends on the distance and the fixed temperature in the central area (Fig. 1).

When heating the LWAs, two steps were differentiated:

– *Prefiring*. In order to determine the effects of the prefiring time on the properties of the LWAs, dry pellets of the *S75F25 mixture*, were subjected to pretreatment in *zone 1* of the rotary kiln in groups of 25 granules, about 20 cm from the entrance. These were maintained there for different dwell times (1.5 min and 5 min), with the temperature in *zone 2* remaining constant (1150 °C). The temperature in the *prefiring area* ranged between 200 °C and 400 °C, depending distance to *zone 2*. This temperature is that corresponds to a fixed temperature of 1150 °C in the *firing area*, in a distance to the entrance of about 20 cm (*prefiring area*) (Fig. 1).
– *Firing*. After the prefiring step, they were put in *zone 2* of the rotary kiln, at 1150 °C, for 3 min and 4 min, depending on the dwell time in the *prefiring area*. In this way, two types of artificial aggregates were obtained:

(i) LWAs prefired for 1.5 min and fired for 3 min. This type of LWAs is hereafter called *LWA-4.5*.