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Microstructural characterisation of lightweight granules made from masonry rubble

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ABSTRACT

The pore structure of lightweight granules made from masonry rubble was studied in order to better understand their engineering properties. Thermally and hydrothermally hardened granules were tested. Analysis by ESEM, mercury porosimetry and sorption methods yield important insight into their microstructure. The thermal granules are characterised by partly melted vitreous areas and large internal macropores that are connected via narrow throats. They show a marginal specific surface area along with a hydrophobic behaviour. In contrast, the hydrothermal granules have an accessible mesoporous system containing plate-like and ink-bottle pores. The shape of their water isotherms depending on the granules' CaO content is sensitive to the morphology of calcium silicate hydrate phases (CSH). The hysteresis changes from a narrow loop that closes at low pressures, which can be attributed to coarser more crystalline CSH, to a large triangular-shaped loop along with a low pressure hysteresis, which is characteristic for fine fibre-like CSH with ink-bottle and plate-like pore morphologies. Granules with fibre-like CSH have the higher specific surfaces areas but those with more crystalline CSH show stronger physisorption of water molecules.

1. Introduction

Lightweight granules are porous and nearly spherical mineral particles with bulk density less than 2000 kg m^{-3}. Today, most commercial lightweight granules are produced by thermal treatment of natural raw materials like clay and shale. They are used in various fields from aggregates for lightweight concretes, insulation bricks and insulation fillings in the construction industry to substrates, filter media and fixed- or fluid-beds for exchange, separation and storage processes in planting and wastewater treatment. Depending on the application, the lightweight granules can consist of single grain or graded particle distributions with particle sizes ranging from a tenth of a millimetre to nearly 100 mm. Whereas both appropriate low intrinsic weight and certain mechanical stability of the lightweight granules are required in most cases, each application has special demands on the microstructural characteristics of the particles. For lightweight concretes, a relatively high total porosity but moderate water absorption arising from well-proportioned fractions of open and closed pores is important [1–4]. Furthermore, textural and chemical characteristics of the granules' surface are important to produce a tight aggregate/cement paste bond [5–7]. For storage and exchange application, a consistent or narrow pore size distribution and pores with a defined pore width determining the water absorption capacity or the ion exchange rate are often crucial factors [8–13].

Expanded clay and shale are produced at burning temperatures up to 1300 °C. In addition to the high energy costs thereby incurred, the availability of suitable raw materials is limited. An alternative is the use of new lightweight granules made from mineral construction and demolition waste. A manufacturing technology has recently been developed in the framework of a German research project [14–18]. Masonry rubble containing, among others, brick material served as raw material. The lightweight granules were obtained in a multistage manufacturing process by thermal or hydrothermal treatment. For thermal hardening, the finely ground rubble was blended with silicon carbide powder (SiC) as foaming agent. The green granules were burnt at temperatures below 1200 °C in a rotary kiln [14–18]. Similar to the manufacturing process of expanded clay or shale, expansion occurs due to gas release (here from the SiC powder) at the same temperature where

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the fine grained rubble material reaches a visco-plastic and partly melted or vitreous state [19–21].

Alternatively, a less energy intensive hydrothermal hardening method under pressurised steam in an autoclave was used [15,16]. This process required the addition of caustic lime powder (CaO) as binding agent. The hydrothermally hardened granules evolve in a process similar to that of the production of calcium silicate masonry units where calcium silicate hydrates (CSH), which are formed by the reaction of hydrated lime and quartz, develop the strength of the aggregates [21,22].

Due to the two different manufacturing methods and varying process parameters, the lightweight granules can be tailor-made. Their bulk densities and particle strengths vary within wide limits between 600 kg m^{-3} and 2000 kg m^{-3} and 1.5 MPa and 13 MPa, respectively [15,23]. To be conscious of strong correlations between engineering properties and microstructural characteristics of the lightweight granules, studies of porosity and pore structure are very important to understand the behaviour of the new lightweight granules in connection with the optimisation of their production process. The present work contributes to these issues by analysing the microstructure of thermally and hydrothermally hardened granules by means of environmental scanning electron microscopy (ESEM), mercury intrusion porosimetry, nitrogen and water vapour sorption measurements for the first time.

2. Experimental

2.1. Materials

Various lightweight granules from thermal hardening (referred to as TH) and from hydrothermal hardening (referred to as HH) were chosen as samples for the microstructural studies. All granules were produced from the same starting material, which was a masonry rubble containing about 50% brick material. The chemical composition of the rubble, which was analysed by means of inductively coupled plasma optical emission spectrometry (ICP-OES) following a total decomposition by microwave assisted acid digestion, is shown in Table 1. In mineralogical terms, the main component of the rubble is quartz followed by calcite [21].

To manufacture the lightweight granules, the heterogeneous masonry rubble was crushed and ground to a powder, which was then homogenised, moistened with water and granulated using a high shear mixer. Afterwards, the green granules were hardened either thermally or hydrothermally. For thermal hardening, 3% SiC powder (the foaming agent) was added and the mixture was burnt at 1180 °C in a rotary kiln [14–18]. The bulk densities of the thermal granules were provided by varying heating regimes and burning times that lead to different expending degrees of the granules. Details are already described in [14–18]. For hydrothermal hardening, 5%, 7% and 9% of CaO powder (the binding agent) was added and the green granules were treated at 200 °C and 1.6 MPa under pressurised steam in an autoclave [15,16]. Due to different amounts of CaO, the bulk densities of the hydrothermal granules varied whereas higher CaO quantities cause lower bulk densities.

The basic engineering properties of the granules are summarised in Table 2. The sample names are indicated by the granules' bulk densities expressed in kg m^{-3}. The sample selection was based on the bulk densities of the granules to cover the whole variety of manufactured materials. All samples meet the criteria of lightweight granules with bulk densities less than 2000 kg m^{-3} and achieve particle strengths higher than 1 MPa, in which the sample strength increases with bulk density. With the similar skeleton density of about 2650 kg m^{-3}, the total porosity of the granules varying between 30% and 80% decreases linearly with growing bulk densities regardless of the hardening method. More details about the engineering properties of the lightweight granules are discussed in [14–18,23].

2.2. Methods

The environmental scanning electron microscopy was performed with a XL 30 ESEM (Philips) to get a first impression of the samples. The ESEM technique enabled the fast investigation of the sample in a low vacuum atmosphere without applying a conductive layer on its surface and further preparation. In all cases, fresh untreated fractured surfaces of the granules were analysed.

To get quantitative information on the microstructure, mercury intrusion porosimetry and gas sorption measurements with nitrogen and water vapour were used. Granules with particle sizes between 4 mm and 8 mm were measured as they were without crushing. Prior to the sample conditioning in each analyser, the granules were dried at 378 K.

The mercury intrusion and extrusion analyses were carried out by an Auto Pore III 9400 porosimeter system (Micromeritics) applying a maximum pressure of 405 MPa according to ISO 15901-1 [27]. Therewith a range of pores and pore entrances,

Table 1
Chemical composition of masonry rubble used for production of lightweight granules.

Component	SiO_2	CaO	MgO	Al_2O_3	Fe_2O_3	TiO_2	SO_3	Na_2O	K_2O	Cl^-	LOI
Amount (% by mass)	68.9	9.2	2.3	12.0	3.7	0.6	0.7	0.7	1.6	0.02	6.9

LOI – loss on ignition.

Table 2
Engineering properties of lightweight granules selected for microstructural studies (TH – thermally hardened, HH – hydrothermally hardened).

Sample (added agent)	Particle strength[a] (MPa)	Bulk density[b] (kg m^{-3})	Skeleton density[c] (kg m^{-3})	Total porosity (%)
TH 640 (3% SiC)	1.1	640	2620	75.6
TH 1160 (3% SiC)	4.9	1160	2670	56.6
TH 1410 (3% SiC)	7.7	1410	2660	47.0
HH 1560 (9% CaO)	6.8	1560	2620	40.5
HH 1700 (7% CaO)	9.9	1700	2670	36.3
HH 1800 (5% CaO)	12.5	1800	2600	30.8
HH 1840 (5% CaO)	12.2	1840	2600	29.2

[a] Determined by modified cylinder test method (volume of test cylinder 200 cm^3) according to EN 13055-1 [24].
[b] Determined by pycnometer method with water according to EN 1097-6 [25].
[c] Determined by helium pycnometry according to DIN 66137-2 [26].