Construction and Building Materials 191 (2018) 176–186

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

# Synthesizing super-hydrophobic ground granulated blast furnace slag to enhance the transport property of lightweight aggregate concrete

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## HIGHLIGHTS

- A super-hydrophobic GGBS is prepared through a ball milling method.
- Optimum synthesis conditions to prepare super-hydrophobic GGBS are explored.
- Applying super-hydrophobic GGBS to lightweight concrete makes it hydrophobic.
- Correlation between the super-hydrophobic GGBS and concrete microstructure is studied.
- Applying super-hydrophobic GGBS to lightweight concrete enhances transport properties.

## ARTICLE INFO

*Article history:*
Received 18 April 2018
Received in revised form 21 September 2018
Accepted 3 October 2018

*Keywords:*
Ground granulated blast furnace slag
Physical carrier
Stearic acid
Hydrophobicity
Lightweight aggregates concrete
Transport properties

## ABSTRACT

This paper studies the feasibility of synthesising super-hydrophobic ground granulated blast furnace slag (GGBS) as water-resisting admixture in lightweight aggregate concrete. The super-hydrophobic GGBS was produced through a ball milling method using the low cost stearic acid. Results show that optimum synthesis involves dry milling stearic acid for 0.5 h with the dosage of 1 wt%, producing a super-hydrophobic GGBS that shows a water contact angle of 155.7°. The morphology, crystalline structure, functional group and chemical state of the atoms were investigated employing SEM, XRD, FTIR and XPS. TEM analysis shows that the thickness of the stearic acid coating is 7.1 nm, confirming the theoretically calculated value. Lightweight aggregate concretes are designed applying an optimized particle packing theory and the effects of super-hydrophobic GGBS on cement hydration, workability, fresh density, strength and transport properties of the concrete are evaluated. Moreover, the relationship between the super-hydrophobic GGBS and the transport properties related performance of the lightweight concrete is discussed. With the addition of the super-hydrophobic GGBS, the capillary water absorption and long-term chloride penetration depth of the LWAC reduce up to about 90%.



## 1. Introduction

The application of lightweight aggregate concrete (LWAC) as both structural and non-structural material has gained much attention [1], and extensive investigations have been carried out on LWAC due to its superior properties like low density, excellent thermal properties and good fire resistance property [2,3]. However, caused by the porous lightweight aggregates (LWA), LWAC has the potential to show high permeability, especially compared to normal density concretes [4,5]. Although the application of lightweight aggregates with relatively closed pores can help to increase the resistance to fluids transport, the rather smooth surface of such aggregates leads to a potential decrease of mechanical strength and the defects on the surface of such aggregates remain passages for the fluids to penetrate [3,6]. Therefore, additional treatments are often needed to enhance the transport properties of LWAC. Nano-materials such as nano-silica have been investigated to refine microstructure of LWAC for better durability performance [5,7,8]. Nevertheless, issues such as the limit to reduce porosity and large water demand due to the high specific surface area, in addition to the high cost, still exist [9,10].

Hydrophobic modification is an efficient way to improve the transport performance by preventing water from penetrating into concrete structures [11]. Most existing strategies for hydrophobic modification for concrete apply extra surface treatment or hydrophobic ingredients in concrete matrix [11,12]. Because of the ease of dispersion and preparation, silane, siloxane or a mixture of these two components are most commonly used for hydrophobic modification for concrete at ambient conditions [13]. However,

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https://doi.org/10.1016/j.conbuildmat.2018.10.018
0950-0618/

because of the relatively high price, their large utilization in concrete is still practically difficult [11,14]. Furthermore, premature cracks can occur as a result of shrinkage of cementitious materials at early age, making the concrete more vulnerable to the ingress of potentially aggressive species, which will significantly decrease the water repellent effect of the surface modification [11,12]. Therefore, the bulk volume modification is a more preferable manner to increase the concrete durability [12].

Compared with the silane and siloxane, saturated fatty acid is much cheaper and has already been used to prepare hydrophobic materials [15,16]. However, a pre-treatment step, such as water bath heating is needed to disperse it as it is in solid state at room temperature and thus it is difficult to get well dispersed during the application process [17,18]. Different from the soft chemical method mentioned above, mechanical approach such as ball milling has also been applied to prepare the super-hydrophobic powders, by which the mechanical coating of hydrophobic agent on the target powders occurs [16,19]. During the milling process, the added chemical agent will be coated on the surface of the powders, consequently endowing them with hydrophobic property. The hydrophobic performance of the powders prepared by this method is dependent on a number of parameters, including such as the nature and amount of the powders and surfactant, ball to solid ratio, milling speed and time [20,21]. Unlike the soft chemical method in which certain parameters crucially affect the preparation efficiency, no significant issues exist in mechanical approaches [20]. Comparing with the soft chemical method with which the target powders are added to the solution of surfactant agent, physical coating method like ball milling sometimes shows coating defects, especially when the powders present irregular shape [21]. The uncoated parts of the target powder will decrease the hydrophobicity. Declan et al. found that the defects of the coating cause the strength decrease of the polymer composite (high density polyethylene based), attributed to the decrease of interfacial adhesion between polyethylene and stearic acid modified peat ash powders [22]. Li et al. reported that high rotation speed and long ball milling time are necessary in order to achieve a perfect coating with ball milling method [23]. Spathi et al. prepared super-hydrophobic powder applying paper sludge ash as the primary material and applied it in concrete to improve the durability performance [16]. However, the preparation process needs long time (8 h) and high stearic acid addition (4 wt%). This may be attributed to the porous structure with significantly high pore volume of the paper sludge ash which absorbs more surfactant and more time is therefore needed to react. Therefore, powders with smaller porosity are desired. Liu et al. successfully prepared hydrophobic stearic acid coated zirconia powders and the BET surface area of $ZrO_2$ is 8.21 $m^2/g$ [24]. With a stearic acid addition of 0.5 wt%, a coating with a very thin layer of about 0.5–0.8 nm can be achieved. Nevertheless, up to now no research has been reported on dealing with a powder with reactivity potential to be a bulk hydrophobic modification agent.

Blast furnace slag is an industrial by-product resulting from ion production, and it consists primarily of silicates, and aluminate and calcium [25]. In order to broaden the application range, the original granules with large-sizes are always grounded to fine particles, known as ground granulated blast furnace slag [25]. The minerals contain melilite, merwinite, dicalcium silicate, wollastonite, anorthite, monticellite, etc. [25,26]. The excellent cementitious property of blast furnace slag has made it a very popular supplementary cementitious material for concrete production [26–28]. In this study, a reactive GGBS powder is used as a carrier of stearic acid, employing a mechanical coating method. It shall be noted that the potential impact coating of GGBS will help to remain the cementitious property of GGBS, which is desired. The optimal synthesis conditions in terms of hydrophobicity are evaluated. The acquired GGBS is assessed by X-ray diffraction (XRD), Fourier transform infrared (FTIR), thermogravimetry analysis (TGA), BET specific surface area and transmission electron microscopy (TEM). The hydrophobic performance is evaluated by the water contact angle measurement. Subsequently the synthesized super-hydrophobic powder is applied to lightweight aggregates concrete to enhance its resistance to fluids transport. One type of natural expanded silicate material is used here as the lightweight aggregates [29]. The influence of the super-hydrophobic slag (H-GGBS) on cement hydration at the early age, flowability, mechanical property, microstructure and hydrophobicity of the lightweight concrete is investigated. More importantly, the developed lightweight concrete is investigated in terms of capillary water absorption and long term natural chloride diffusion concerning the durability aspects and the effect of the super-hydrophobic slag powder is discussed.

## 2. Experiment

### 2.1. Materials and mix design of LWAC

#### 2.1.1. Materials

The ground granulated blast furnace slag (GGBS) was supplied by ENCI B.V. (the Netherlands). The elemental composition of the GGBS is determined by X-ray fluorescence, as: 34.61% $SiO_2$, 37.63% CaO, 13.26% $Al_2O_3$, 9.94% MgO, 0.47% $Fe_2O_3$, 1.24% $SO_3$, 0.47% $K_2O$, 0.98% $TiO_2$, 0.01% Cl, and 0.46% L.O.I. The SEM morphology of the raw GGBS and milled GGBS is investigated by a Phenom Pro analyser and are shown in Fig. 1 (a) and (b). CEM III/A 52.5 N (supplied by ENCI B.V., The Netherlands) was used, considering both the sustainability and durability performance by the contained slag.

Four size fractions of natural expanded silicate material were used as lightweight aggregates which are supplied by ROTEC GmbH & Co. KG Rohstoff-Technik (Germany), including 0.09–0.3 mm, 0.5–1.0 mm, 1.0–2.0 mm and 2.0–4.0 mm (see Fig. 2). The aggregates have a very low thermal conductivity of about 0.08 W/(m·K) with a crushing strength from 12 $N/mm^2$ (fraction 2–4 mm) to 22 $N/mm^2$ (fraction 0.09–0.3 mm). A polycarboxylate ether based superplasticizer (SP) was used to adjust the flowability of the designed concrete to the desired value. A stearic acid (reagent grade, 95%) was used to synthesize the super-hydrophobic GGBS.

#### 2.1.2. Synthesis of the super-hydrophobic slag

Ball milling method was used to prepare the hydrophobic GGBS powder with stearic acid, using a porcelain ball mill pot (Fritsch; Pulverisette 5) loaded with 20 alumina milling balls (d = 20 mm) inside. The stearic acid and GGBS were added together into the milling pot. Different experimental conditions including the milling time, speed and stearic acid dosage were experimented to achieve the optimum performance, in terms of water-contact angle that is determined with a pressed pellet. The influences of rotation speed and time to the hydrophobic performance of GGBS were investigated. The influence of stearic acid dosage on the hydrophobicity of the GGBS powders was studied by using 0.5, 1, 2 and 4 wt% additions of stearic acid with all other processing variables kept constant.

#### 2.1.3. Mix proportions of the LWAC

As aforementioned, the bulk treatment was carried out by replacing CEM III/A 52.5 N with the S-GGBS. Reference samples were prepared for comparing the effect of S-GGBS incorporation on the performance of the designed LWAC. The modified Andreasen and Andersen model was applied for determining the mix pro-