Cement and Concrete Research 115 (2019) 59–69

Contents lists available at ScienceDirect

Cement and Concrete Research

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

Application of an alternative way for silica fume dispersion in cement pastes without ultrasonication

Eirini-Chysanthi Tsardaka, Maria Stefanidou*

Laboratory of Building Materials, Dept. of Civil Engineer, AUTH, Greece

ARTICLE INFO

Keywords:
Silica particles dispersion
Sodium bicarbonate
pH
Cement pastes
Ultrasonication

ABSTRACT

The dispersity of silica particles affects their influence in a composite. In this work, a solution for silica fume (SF) dispersion is proposed for application in cement mixtures, as de-agglomeration method. Sodium bicarbonate salt solution 0.01 M pH 13, with addition of SF consist the proposed dispersion (SBCS). The solution has low cost it is stable, easy and quick to be prepared and does not require special equipment. Dispersion was characterized using particle size distribution and ATR. Cement pastes with ultrasonicated colloidal suspension of SF in water (US) and with sodium bicarbonate - SF (CDS) are compared. DTA-TG, XRD analysis and SEM observation, along with the determination of mechanical and physical properties of the produced specimens are reclaimed. SBCS gave illustrative results after subjecting the specimens in durability tests. The results indicated that SBCS can be used to disperse SF affecting positively the microstructure and the macro-properties of cement systems.

1. Introduction

Nanocomposites are an extensive research subject, due to their contribution in several scientific fields. They have been used as useful tools in material and engineering sciences. Silica fume is the most studied additive that successfully enhances the mechanical properties of cement and contributes to its hydration [1]. Silica particles have been proved that consume calcium hydroxide that is produced during the hydration of C_3S of cement and contributes to higher early strength [2–4]. One of the main problems of nano-particles is their tendency to agglomerate, due to their high surface energy [5,6]. According to Hartley et al., the particle size of powder has been inversely correlated to cohesiveness. The research team pointed out the “breakdown of agglomerates into primary particles” as a main dispersion mechanism stage [6].

Colloidal suspensions of silica particles have been widely studied for their various properties. For instance, the absorption of polymeric compounds has demonstrated certain behavior depending on temperature, polymer used and stability obtained [7–9].

The most wide-spread and accepted way to incorporate silica particles in composites is to prepare a colloidal suspension, by adding silica in aqueous medium and ultrasonicate the solution for defined time. The definition of time plays an important role to the final dispersion. An extensive duration of sonication may lower the size of agglomerates or lead to re-agglomeration, depending on the conditions of the system. So, the use of ultrasonic is a double side issue. Some methodologies, in order to avoid or reduce agglomeration, proposed that sonication time [10] and the procedure of dispersion production influence the dispersion itself and the resulted mixtures as well. Mandzy et al. indicated that sonication time affects the polydispersity index of the dispersion and that 30 W of power and frequency of 20 kHz for 15 s start to break agglomerates effectively [11]. According to Jafari et al. long-time sonication does not necessary mean better dispersion as it can cause agglomeration to a greater extent than short-time sonication [10]. Nevertheless, for ultrasonication of silica in aqueous mediums, 60 minute time has prevailed in literature and has been applied in cement pastes [12].

Ultrasonic and stirring at the same time is a methodology that is often used for the incorporation of nanoparticles in cement-based materials [13]. This is a methodology that inserts one more step in the procedure of the specimen preparation and plays an important role in the action of SF, as it affects the agglomeration behavior. Moreover, ultrasonication is time-consuming procedure and requires specific instrumentation that can be used only in laboratory and if this procedure will perform in large (industrial) scale, the cost could be a deterring factor. Another considerable factor is how practical is to apply the sonication procedure, especially in high quantities of concrete. The idea of a low cost and easy to prepare solution that could disperse silica nanoparticles for application in building materials is appealing. Using compensatory dispersed SF, which means homogenous suspension,

* Corresponding author.
E-mail address: stefan@civil.auth.gr (M. Stefanidou).

https://doi.org/10.1016/j.cemconres.2018.10.002
Received 19 January 2017; Received in revised form 5 October 2018; Accepted 9 October 2018

leads to denser structure and increases the workability of concrete [1]. Plus, it has been proved that the addition of superplasticizer after the sonication process increased the compressive strength, comparing to the addition of superplasticizer before ultrasonication [14].

According to literature, silica particles have also been dispersed in aqueous mediums other than pure water [15,16,19]. In particular, the presence of sodium chloride (NaCl) in silica suspension was found to be an improving agent of stability for the suspension of colloidal silica. Moreover, Terpilovski et al. indicated that in water and 0.1 M NaCl solution, nanosilica particles were "electrostatically stabilized" at pH values lower than 8.7 and 10.6 respectively [16,18]. The agglomerates though were still formed and reported that were sedimented to the bottom of the container. Additionally, other researchers have commented that as the concentration of electrolyte (NaCl) increases, so does the viscosity of the dispersion [15]. Plus, concerning the stability study they performed, NaCl lead to a small increase of negative zeta potential [15]. These research studies concluded to useful results for suspension of SF, but sodium chloride cannot be used in a dispersion that is about to be applied in building materials, because of chloride ions that lead to deterioration. Chloride anions tend to bond with metallic cations, especially calcium (Ca^{2+}) and form hazardous salts. Moreover, chloride anions corrode the reinforcement of concrete and that is why there are very low limits in the chloride anions content in relevant regulations ($Cl^- < 0.1$ wt%) [20].

This research is an effort to overcome the ultrasonic process by dispersing SF in an inorganic aqueous medium, such as bicarbonate salts, in order to apply the improved dispersion in cement-based materials. The non-sonicated dispersion of SF in inorganic salts and the conventional methodology that gives ultrasonicated SF in water will be compared. An electrolyte that cannot form corrosive salts in cement based materials was chosen, in order to disperse SF and cannot prevent cement corruption. Sodium bicarbonate salts with adjusted pH will be studied as a medium to disperse SF and avoid the formation of corrosive salts, which are given in literature, such as chloride formations, for building materials applications [21]. Also, carbonate anions can be captured in the structure without interfering to the hydration of cement. SBCS was tested and characterized, using particle size analysis and (Attenuated Total Reflectance) ATR spectroscopy. The results were evaluated and discussed for the selection of the proper pH value and concentration of solution, for the dispersion of SF. The effects of SBCS in cement pastes were tested using Differential Thermal Analysis-Thermogravimetric Analysis (DTA-TG), X-Ray Diffraction analysis and SEM-EDS instrumentation in order to determine structural and mineralogical changes in the specimens. Additionally, the mechanical and physical properties, such as porosity and capillary absorption of the produced cement specimens have been reported. Finally, the resistance of the produced pastes in ageing factors (freeze-thaw cycles, sea water and sodium sulfate cycles) was evaluated.

2. Experimental

2.1. Preparation of dispersion

For the preparation of sodium bicarbonate salts, a quantity of sodium bicarbonate ($NaHCO_3$) was diluted in distilled water and 0.01 M solution was prepared. The pH value of the prepared solution was pH 8.5 and in order to adjust the pH from 10 to 13.5, drops of NaOH 2 M were added under stirring. The pH values were recorded with a pH-meter. The alkaline pH values were planned in order to simulate the environment of cement.

An amount of 10 mL of sodium bicarbonate salts at different pH values (pH 10, 11, 12, 13 and 13.5) was transferred in a small coded glass-holder and a weighted quantity of SF was added in the holder. Silica particles were tested in percentages of 1.5% and 3% w/v of the total amount of the solution, with addition of 0.15 ± 0.001 g and 0.3 ± 0.0001 g SF respectively. The urn was sealed, shacked instantly manually and left aside for 4 to 6 h without any use of mixing or ultrasonic assistance. It is worth to be mentioned that SBCS above pH 13.0 became pellucid in 6 h, which is in agreement to the literature [22,23]. From pH 13.0 and below, the dispersion had a white nuance, which was lighter at pH 13 than that at pH 10.

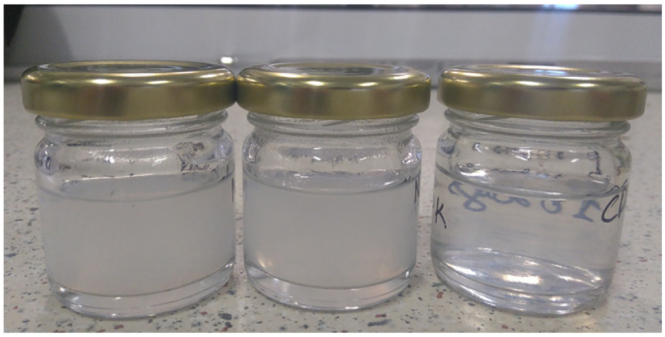

Fig. 1. SBCS with 1.5% SF at pH 11 (left), pH 12 (middle) and pH 13.0 (right).

For reference dispersion samples the following solutions with 1.5% w/v and 3% w/v SF were prepared: 10 mL of distilled water (pH 5.81) with addition of 0.15 ± 0.001 g and 0.3 ± 0.0001 g SF respectively and 1 hour sonication, and 10 mL of sodium bicarbonate (pH 8.5) with SF at same proportions, 1.5% and 3.0%. In both cases of reference dispersions, they appeared thicker white than the dispersion of SBCS at pH 10–13. The SBCS above pH 13.0 was transparent (Fig. 1). The content of these vials was tested after one day by ATR analysis. The same procedure for sample and reference dispersions was repeated for 100 mL total amount of dispersion for the particle size analysis test.

2.2. Materials and instrumentation

Sodium bicarbonate ($NaHCO_3$) was supplied from Sigma-Aldrich and sodium hydroxide (NaOH) from Panreac. Silica particles were hydrophilic and amorphous, N20 from Wacker and had BET surface 175–225 m^2/g.

For the characterization of ultrasonicated dispersion a particle size analyser Mastersizer 2000 of Malvern Instruments (wet phase) and an ATR analyser Alpha- Platinum ATR of Brucker Company, were used. For the ultrasonication of sample contained water and SF, Mastersizer 2000 was used with pump speed 3000 rpm, 40 W power and frequency 40 kHz.

Analytical balances, Kern PCB 4000-2 (max capacity 4000 ± 0.01 g) was used to weight cement and B204-S/FACT of Mettler Toledo (max. capacity 220.0 ± 0.0001 g) was used to weight silica particles and sodium bicarbonate quantity. For the preparation of the cement specimens a mixer, a Vicat apparatus and metallic molds of (25 × 25 × 100) mm and (25 × 25 × 50) mm were used. The mixer was automatic programmable mixer Automix Controls.

For the evaluation of the produced pastes the following instruments were used. A Simultaneous DTA-TG (Differential Thermal-Thermogravimetric Analysis), SDT 2960 TA Instruments, was used for the determination of calcium hydroxide ($Ca(OH)_2$) and calcium carbonate ($CaCO_3$), under N_2 atmosphere from 10 to 1000 °C. X-Ray Diffraction analysis was performed by PW 1840 Phillips diffractometer. Flexural and compressive strength, were determined using a Technik ToniNorm device that functions according to EN 196-1 [17]. For the microstructural characterization of the specimens, the SEM-EDS instrument used was JEOL JSM-840A Scanning Electron Microscope. Porosity was measured according to RILEM CPC11.3 method of water absorption under vacuum [25].

2.3. Characterization of raw materials

Cement CEMII32.5N was used. The characteristics of cement, such