Materials and Design 90 (2016) 29–37

Contents lists available at ScienceDirect

Materials and Design

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

The sequel of modified fly ashes using high energy ball milling on mechanical performance of substituted past cement

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ARTICLE INFO

Article history:
Received 13 May 2015
Received in revised form 17 October 2015
Accepted 20 October 2015
Available online 21 October 2015

Keywords:
Ball milling
Fly ash substitution
Structural analysis
Compressive strength
Cement paste

ABSTRACT

The consideration of planetary ball mill processing is attempted to obtain modified fly ashes powders that are used as substitutes in a cement paste. Ball milling conditions are related to structural modifications of processed powders. These modifications are retrieved from X-ray diffraction, infrared spectroscopy, powder granulometry, and scanning electron microscopy. As an industrial application, the milled fly ashes are substituted to two different cements to show possible effect on compressive strength of cement pastes. Structural analysis of milled fly ashes show loss of crystallinity of the magnetite phase, nanosizing trend of quartz and mullite phases with some straining that takes place after only 1 h of milling. The drop of particle size of the milled fly ashes is also evidenced but with tendency to agglomeration and particle shape modification from spherical to irregular forms. Mechanical performance results related to modified cement pastes indicate successful substitution of cements by milled fly ashes up to 50%. Substitution results in levels of compressive strength as large as 70 MPa for particular combination of milling time, fly ash content and curing time.

1. Introduction

Fly ashes are widely used as cement substitutes inasmuch as they show major technical benefits, including reduced hydration heat and permeability, improved resistance to alkali silica reaction, sulphate attack, delayed ettringite formation, and external sulphate attack [1,2]. The imputed performance of fly ashes sounds with applications like road construction [3,4], waste and soil treatment [5–8]. Fly ash is a predominantly inorganic residue obtained from the flue gases of furnaces at pulverized coal power plants [1,9,10]. Fly ash is generally produced at 1200–1700 °C from various inorganic and organic constituents of the parent coal. The powder is collected after rapid cooling from high temperature to nearly 200 °C [11,12]. The characteristics of produced fly ash differ depending on the coal source, the method of combustion of power plants and typologies of emission control devices, storage and handling [12]. As per the ASTM C618-12a guideline, the fly ash containing more than 70% of SiO_2, Al_2O_3 and Fe_2O_3 is classified as Class F type fly ash. If the fly ash composition consists mainly of silica, alumina and calcium containing SiO_2, Al_2O_3 and Fe_2O_3 minimum up to 50%, this is referred as Class C fly ash [7,12–14]. Fly ash particles are very fine, mostly spherical and vary in diameter. Generally, they look like tiny solidified bubbles or spheres of various sizes. The average particle size is about 10 μm but can vary from less than 1 μm to over 150 μm [7]. Therefore, fly ash shows a wide variation in its physicochemical and mineralogical properties. The main detected crystalline phases in fly ash are mullite, quartz, magnetite, haematite and anhydrite [7,9,10,15]. Fly ash is a pozzolanic material formed by finely-divided amorphous alumino-silicate with varying amounts of calcium. Fly ash mixture with Portland cement and water induces reaction with the calcium hydroxide released by the hydration of Portland cement to produce various calcium-silicate hydrates (C-S-H) and calcium-aluminate hydrates (C-A-H) [16]. Some fly ashes with higher amounts of calcium also display cementitious behaviour by reacting with water to produce hydrates in the absence of a source of calcium hydroxide. These pozzolanic reactions are beneficial to the concrete in the way that they increase the quantity of the cementitious binder phase (C-S-H) and, to a lesser extent, calcium-aluminate hydrates, improving the long term strength and reducing the permeability of the system. Both of these mechanisms enhance the durability of the concrete [14,17–20]. However, there are several issues to resolve before using fly ashes in cement mixtures. The main problem lies in the insufficient activity of the fly ashes particles during the hydration reactions, which are important to establish the mechanical characteristics of cement. Pozzolanic reactions are, in comparison to the hydration reactions of Portland cement, much slower

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http://dx.doi.org/10.1016/j.matdes.2015.10.109
0264-1275/

and they occur to an observable extent only after 7 days to 28 days [14,17–20]. For these reasons, several contributors proposed different methods for fly ash activation based on alkali-activation [21–24], curing temperature effect [18], vitrified fly ash [25], quicklime active fly ash [26] and mechanical activation [6,27]. One reported work by Kocak et al. [28] proves that the use of fly ashes without activation decreases the compressive strength of blended cements for any fly ashes content in the range (5–25) % compared to the reference value. However, the authors highlight improved mechanical strength even below the reference indicating activation of pozzolonic reactions at large curing time [28].

Elsayed [2] confirms the negative effect of fly ash substitution on water permeability and strength of concrete. The compressive strength of modified concrete deteriorates with cement replacement by fly ash and reaches even 20% of missing strength with 30% of fly ash substitution.

The negative effect of inactivated fly ashes is generally compensated by high calcium content. This may raise the compressive strength of the modified concrete to levels that are 25% larger than the control concrete strength [15–17].

Fly ashes activation is attempted by several contributors. Antiohos et al. [26] report a positive influence of quicklime addition on the mechanical properties and hydration degree of blended cements containing different fly ashes. The authors [26] show improvement of strength of the activated mortar by 5% after 90 curing days. Dyer and Dhir [25] report also another way of fly activation using vitrification process. Their result demonstrates a more contrasted result on compressive strength of modified mortars depending on fly ash content and curing duration. In this study, the cement is substitution by fly ash with a proportion reaching 40% and for those formulations exhibiting positive tendencies; the compressive strength is only comparable to the control strength. High energy milling is also considered as an efficient way to activate fly ashes. This route allows in-depth modification of fly ashes including structure shift, particle size reduction, particle shape alteration and increase in the specific surface [10,29]. For instance, ball milling is attempted by Chen et al. [27] to treat fly ashes from municipal solid waste incinerator. The authors demonstrate that the replacement of less than 10% of the cement by these processed fly ashes induces slight improvement of the past cement strength. Krishnaraj et al. [30] have studied the effect of replacement of ordinary Portland cement with weight content up to 60% wt. of raw and milled (ultra-fine) fly ashes in mortar confection. The authors show [30] that replacement of the cement by an amount larger than 15 wt.% result in an increase of the compressive strength at only 28 curing days if milled fly ashes are used as substitutes instead of raw fly ashes.

Within the same line of thought, this work aims at revealing the effect high energy ball milling conditions on the structure and thermal properties of fly ashes. The potential use of milled fly ashes is tested on modified cement pastes through the substitution of two different cement grades. The substitution of CEM I, which contains more than 95% of clinker, is intended to decrease CO_2 negative impact of civil engineering materials on environment. CEM II is also used to possibly promote pozzolanic reactions since this grade contains slag (more than 22%). These processed fly ashes are found to promote the mechanical performance of modified past cements under a controlled amount of cement substitution.

2. Materials and methods

The cements used in this study are CEM I 52.5 and CEM II/B-M (S-LL) 32.5 R CP1 NF EN 197-1 from Calcia. Fly ashes are provided by Eiffage Company. The chemical composition of both unmilled and milled fly ashes is performed using X-ray fluorescence spectroscopy. These experiments are carried out using the S2 RANGER instrument from Bruker.

The milling of fly ash powder is achieved using a planetary high-energy ball mill (Retsch PM 400). The planetary high energy ball mill is composed of four vials mounted on a planar disc. With the rotation of the disc, the vials move in a circular and in opposite direction compared to the disc rotation. The rotation speeds of the disc and the vials are $\Omega = 400$ rpm and $\omega = 800$ rpm, respectively. The milling time t is varied in the range (1–9) hours. Thirty-millimetre diameter steel balls and 500 ml volume steel vials are used. The weight of powder samples is adjusted to 200 g per vial. The ball-to-powder weight ratio is 4. We mention that vials of planetary ball mill (RETSCH PM 400) are adequately sealed to avoid air contamination during milling process. Also, in order to avoid iron or chromium contamination of the processed powder by elements from balls or vials, a delay of 30 min is imposed after 1 h of milling.

The reference past cement is prepared by mixture of cement (C) and water (W) with W/C content ratio equalling 0.3. Modified paste cement by milled fly ash substitution is prepared as follows: for CEM I 52.5 cement grade, substitution by 50 wt.% by as-provided and milled fly ash is attempted up to milling time of 3 h. For CEM II 32.5 cement grade, lower substitution is considered by two levels (15 wt.% and 30 wt.%) and a full range of milling time including 1, 3, 6 and 9 h.

Mixture of cement + fly ash and water is kept constant with ratio (W/C + fly ash) equal 0.3 for both cement grades. Paste mixtures are poured into right-prism moulds of dimensions $4 \times 4 \times 16$ cm^3. Samples are unmould after 24 h, cut into 4 cubes of $4 \times 4 \times 4$ cm^3 and stored in water vats at room temperature.

X-ray investigation is performed using Bruker D2 phaser diffractometer with a continuous scanning mode and Cu Kα radiation ($\lambda = 0.1541$ nm). The lines are measured in the 2θ range (5–100) ° by an increment of 0.02° for 15 s. The software used for building the X-ray diffraction diagrams is DIFFRAC.EVA with ICDD PDF2. In order to improve the relevance of the time evolutions of X-ray diffraction diagrams, X-ray investigation is conduced according to different milling durations from the same vial sampling. The X-ray patterns of fresh fly ashes powders are repeated four times. The change in lattice parameters is calculated from the fitting of the X-ray patterns using Winnel software. This fitting considers the shift of the high angle diffraction line for all X-ray patterns using Bragg's law equation:

$$2dsin(\theta) = n\lambda \tag{1}$$

where d is the spacing between the planes in the atomic lattice; θ is the angle between the incident ray and the scattering planes; n is an integer determined by the order given (in our case $n = 1$); λ is the wavelength of incident wave.

For quartz structure, the average lattice parameter a and c are calculated for all X-ray patterns from different angles at lines (100), (101), (201) and (112) from the expression:

$$\frac{1}{d^2} = \frac{4}{3}\left(\frac{h^2 + hk + k^2}{a^2}\right) + \frac{l^2}{c^2} \tag{2}$$

with (hkl) are Miller indices related to different angles; a and c are lattice parameters.

For mullite orthorhombic structure, the average lattice parameter a, b and c are calculated for all X-ray patterns from different angles at lines (110), (210), (001) and (121) using the following expression

$$\frac{1}{d^2} = \frac{h^2}{a^2} + \frac{k^2}{b^2} + \frac{l^2}{c^2}. \tag{3}$$

The accuracy related to the above expressions is evaluated using the standard deviation for all lattice parameters. The analysis of all X-ray patterns shows that the accuracy of lattice parameter evaluation is below 3×10^{-4} nm.