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Influence of planetary ball milling parameters on the mechano-chemical activation of fly ash

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ABSTRACT

This study illustrates the design of statistical analysis by Taguchi methodology to obtain nanostructured fly ash by planetary ball milling. An orthogonal array and analysis of variance were employed to analyze the effect of milling parameters. A class-F fly ash was subjected to planetary ball milling induced mechano-chemical activation aided by a surfactant. Ball milling parameters, such as ball-to-powder weight ratio, type and quantity of surfactant and type of medium were varied as guided by the Taguchi design. The nanostructured fly ash was characterized by dynamic light scattering, BET surface area analysis, X-ray diffraction, FTIR spectroscopy, scanning electron microscopy, field emission scanning electron microscopy and transmission electron microscopy. The ball-to-powder weight ratio and the surfactant type are the major influencing factors on lower crystallite size and average particle size and higher specific surface area. The surface modification of fly ash was confirmed by FTIR spectroscopy. The nano fly ash produced by this method has a wide application potential in polymer industries as reinforcement in composites.

1. Introduction

Fly ash (FA) is a by-product of thermal power plants, which is obtained in huge quantities upon combustion of pulverized coal in the coal-fired furnaces. Though in the last four decades various alternate energy sources have come into the limelight, the hyperbolic use of coal as a prime energy source cannot be counterbalanced. The environmental impact of FA in terms of its massive generation, large usage of land for disposal, and the impact on surrounding areas are well known [1]. In addition, FA could also affect human health through direct inhalation or ingestion of airborne or settled ash [2]. Characterization of FA in terms of composition, mineralogy, glass content, surface chemistry and reactivity is of fundamental importance in the development of various applications of FA. The potential applications of FA are: soil amelioration agent in agriculture, in the manufacturing of glass and ceramics, in the synthesis of geopolymers, as catalysts and catalyst supports, as an adsorbent for gases and waste water processes, and for the extraction of metals [3,4]. The principal components of FA are silica, alumina, ferrous oxide, and calcium oxide with varying amounts of carbon. The American Society for Testing and Materials (ASTM) groups FA into two classes: C and F. Class-F FA has a total oxide content (of Si, Ca and Fe) of greater than 70%, while it is less than 70% in the Class-C variety [5]. The morphology of FA particles is controlled primarily by the combustion temperature and the subsequent cooling rate. Homogeneous condensation of the flue gases results in FA particles of size in the range of 0.2 to 10 μm. The excluded mineral matter undergoes a series of complex transformations to form predominantly spherical particles in the size range 10–90 μm [6].

Pre-processing of FA involving 'mechano-chemical activation' refers to enhancing its reactivity through combined effects of decreased crystallite size and physicochemical changes induced in the bulk as well as on the surface through high energy planetary ball milling [7]. There are different types of ball milling methods based on the movement of milling balls and vial, such as vibration mill, planetary mill and attritor. In the case of planetary ball milling, the main factors that affect the mechanical activation include rotation speed, size of balls, weight ratio of balls to powder, medium of milling, milling time and extent of filling the vial [8]. During mechano-chemical activation of FA there is uniform mixing of all the charge materials in the milling vial accompanied by repeated fracture, cold welding and deformation of particles. The particles undergo severe plastic deformation due to their collision with the balls and entrapment between the inner walls of the vial and the balls [9,10].

Design of experiments is the process of planning, designing and analyzing experiments. It is necessary to integrate a simple and powerful statistical method into the experimental design methodology to draw conclusions effectively and efficiently [11]. The Taguchi technique provides a simple, efficient and systematic approach to determine the influence of parameters in a manufacturing process. The parameter design procedure determines the factor levels that can generate the best performance of the product or process under study. And it is used to study the entire parameters' space with only a small number of experiments [12,13]. In general, the Taguchi method uses an "orthogonal

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array" of design of experiments with defined logical procedure or algorithm implementation to analyze the experimental data to find the influence of the parameters. Moreover, an analysis of variance (ANOVA) is employed to estimate the error variance and determine the percentage contribution of the individual factors [14,15].

Earlier, Patil et al. [16] improved the compatibility between ethylene–octene copolymer (EOC) and FA by modifying the latter by mechano-chemical treatment. Similarly, the nanostructured FA obtained by mechano-chemical activation was incorporated into a biodegradable poly(vinyl alcohol) matrix by solution mixing aided by ultrasonication. The incorporation of a very small amount of the nanostructured FA led to a substantial increase in crystallinity of the polymer matrix. The improvement in physico-mechanical properties of these composites is encouraging as this strategy could help eliminate environmental pollution due to FA in a profitable manner [17].

In this study, the statistical Taguchi design method with four factors and three levels (3^4) of L_9 orthogonal array was used to study the influence of experimental parameters in the preparation of mechano-chemically activated FA (MCA-FA). The various factors considered during mechano-chemical activation of FA were: change in surfactant type, quantity of surfactant, types of medium and weight ratio of ball-to-powder. The MCA-FA was characterized by X-ray diffraction (XRD), BET surface area analysis, dynamic light scattering (DLS), FTIR spectroscopy, scanning electron microscopy (SEM), field emission scanning electron microscopy (FESEM) and transmission electron microscopy (TEM). The effects of factors on the response mean and variation were estimated by the Taguchi orthogonal array. The response mean is the effect of an independent variable on a dependent variable by averaging across the levels of any other independent variables. Here each factor can be assessed independently of all the other factors, so the effect of one factor does not affect the estimation of a different factor. Statistical analysis of variance (ANOVA) was also employed to decide the effect of significance of the input factors.

2. Materials and methods

2.1. Materials

FA was collected from Raichur (NTPC) Thermal Power Station, Karnataka, India. Analytical grade surfactants, sodium lauryl sulfate (*M.wt.* - 288.38) (SLS), N-cetyl-N,N,N-trimethyl ammonium bromide (M.wt. -364.46) (CTAB) and Triton X-100 (*M.wt.* - 646.87) (TX-100) were procured from Sisco Research Laboratories, Mumbai, India. Toluene, ethanol and ethyl acetate were obtained from Nice Chemicals Private Limited, Cochin, India, which had purities in excess of 99%, and were used without further purification.

2.2. Pre-treatment of fly ash

As received FA was washed with distilled water and the carbon that creamed up during washing was removed. It was then dried at 100 °C for 5 h to remove water. This dried FA (called as fresh FA) was sieved using British Standard Sieves (BSS). Fresh FA fractions that passed through mesh no. 170, and got retained on mesh no. 200 were collected and magnetic separation was carried out manually to remove the magnetic impurities. The resultant FA was mechano-chemically activated subsequently.

2.3. Experimental design: selection of factors and their levels

The most important stage in the design of an experiment lies in the selection of control factors. Taguchi created a standard orthogonal array to accommodate the various factors and levels in the design. The experimental design chosen for mechano-chemical activation of FA with four factors and three levels (3^4) is shown in Table 1. Taguchi's L_9 orthogonal array was used to achieve the control factors, which affect the output response, such as crystallite size, average particle size and specific surface area (Table 2). Each row in the table represents a trial condition with the factor levels. The columns correspond to the factors specified in this study and each column contains three levels.

Table 1
Assignment of factors and levels of L_9 (3^4) orthogonal array.

Levels	Factors			
	A	B	C	D
	Surfactant type	Surfactant quantity (wt.%)	Medium	Ball to powder ratio
1	Anionic (SLS)	2	Toluene (T)	10:1
2	Cationic (CTAB)	4	Ethanol (E)	12:1
3	Non-ionic (Triton X-100)	6	Ethyl acetate (EA)	8:1

Anionic — sodium lauryl sulfate (SLS).
Cationic — N-cetyl-N, N, N-trimethyl ammonium bromide (CTAB).
Non-ionic — Triton X-100 (poly oxyethylene octyl phenyl ether).

2.4. Mechano-chemical activation of FA

Mechano-chemical activation of FA was achieved by using a high energy planetary ball mill (PM 100; Retsch, Germany). FA was charged into a 250 mL tungsten carbide lined vial of the planetary carrier and tungsten carbide balls of 10 mm diameter were used for milling the FA powder. The milling was carried out for 48 h at 250 rpm. The optimum rotational speed of the mill was determined using Eq. (1) proposed by Rose and Sullivan [18,19].

$$N_c = \frac{1}{2\pi}\sqrt{\frac{g}{R\sqrt{1-\alpha}}} \tag{1}$$

where N_c is the critical rotation speed of the ball mill, g is the acceleration due to gravity, R is the inner radius of a vial and α is the volumetric filling ratio of the particles.

For every 2 h of continuous milling, a break time of 60 s was given at an interval of 30 min to avoid heat build-up. The mechano-chemically activated FA will be mentioned as MCA-FA hereafter.

2.5. Characterization MCA-FA

2.5.1. XRF spectroscopy

The elemental compositions of fresh FA, sieved FA and magnetic separated FA were determined by an X-ray fluorescence (XRF) spectrometer (Axios^{mAX} PANalytical, Netherlands) using glass discs prepared by fusing the FA sample with lithium tetraborate.

2.5.2. X-ray diffraction

XRD measurements were carried out to find the crystallite size of MCA-FA, with the help of a Goniometer (JEOL DX-GE-2P, Japan) using CuK_α radiation ($\lambda = 1.542$ Å) at an accelerating voltage of 30 kV and a current of 20 mA. The samples were scanned at a speed of $1°\cdot min^{-1}$ in the 2θ range of 10–90°.

2.5.3. Morphology studies

A field emission SEM (FESEM) (LEO SUPRA55, Carl Zeiss, Germany) and SEM (JEOL-JSM-6380LA, Japan) were used to evaluate the texture and morphology of fresh FA and MCA-FA. The samples for FESEM were prepared by the dip coating of a dilute solution of FA and MCA-FA in ethyl-acetate on a silicon wafer. Prior to FESEM analysis, FA was sputtered with gold in a sputtering unit (JEOL JFC 1600, auto fine coater, USA). The images were taken at suitable accelerating voltages for the best possible resolution using secondary electron imaging. Transmission electron microscope (TEM) (JEOL, JEM-2100, Japan) images were