Study on coal fly ash classified by bulk density

Shuxia Feng* and Yanqi Li
School of Architectural Engineering, Shandong Yingcai University, Jinan, China

*Corresponding author e-mail: fengshuxia1982@126.com

Abstract. As a major country with coal as its main energy source, China has great pressure on the production and reuse of coal fly ash. In this paper, three kinds of original low calcium fly ash from different regions were divided into five bulk density particle groups, < 1 g/cm³, 1-2 g/cm³, 2-2.5 g/cm³, 2.5-2.89 g/cm³ and > 2.98 g/cm³. The results showed that the bulk density of fly ash particles was mostly concentrated in 1-2.5 g/cm³. There were significant differences in the chemical composition and activity of the particle groups with different bulk density, but no consistent changes were observed. The correlation degree between acid oxide content and activity was significantly higher than that between bulk density and activity, indicating that acid oxide content had a significantly higher influence on activity than bulk density.

1. Introduction

Coal fly ash is a kind of solid waste that generated after coal burning. The particles of fly ash have significant differences in bulk density, size, composition, activity and morphology. In addition, the above characteristics of fly ash from different sources or batches are also different due to the different coal burning types, minerals contained in coal, combustion conditions, and collection technologies. Therefore, it is an effective way for utilization of fly ash by classifying fly ash particles and applying them according to their characteristics.

Presently, fly ash were mostly classified according to particle size by sieving. Previous studies have shown that particle size has a significant impact on the chemical composition, morphology, vitreous content and pozzolanic activity of fly ash [1-2]. With the decrease of particle size, the content of regular spherical microbeads increased, the content of irregular lumpy particles decreased [3], the content of modifier (CaO+MgO+Na₂O+K₂O) in the particles increased, and the content of Fe₂O₃ decreased [4]. Generally, the greater the fineness of fly ash, the higher the pozzolanic activity, and the higher the strength of mortar or concrete prepared by fly ash [5-6]. The bulk density of fly ash from different sources without any classified is quite different, most of which are between 1.9-2.9 g/cm³. Due to the different composition and structure of fly ash particles, bulk density among each particle is also different. There are floating bead with bulk density less than 1 g/cm³ and iron beads with bulk density greater than 3 g/cm³. There were relationships between bulk density and various characteristics of fly ash. Hemmings and Berry [7-8] studied the relationship between particle bulk density and chemical composition of fly ash. The variation law between particle morphology and bulk density was also introduced in another literature [9]. According to the review, the existing literature focused on particle size classification. There was a little research on bulk density classification of fly ash, and only the correspondence between bulk density and chemical composition has been studied. There was no report about the relationship between bulk density and pozzolanic activity of fly ash.

In this paper, original fly ash was classified into five bulk density ranges using heavy liquid and
water as sorting medium. The effect of bulk density on chemical composition and activity of fly ash was studied in order to provide basic data for bulk density classification of low-calcium fly ash.

2. Materials and methods

2.1. Materials

Three types of low calcium coal fly ash from power plant in different regions were used in this experiment, which were denoted as F1, F2 and F3 respectively. The chemical composition of original fly ash was shown in table 1.

| Composition | CaO | SiO₂ | Al₂O₃ | Fe₂O₃ | SO₃ | TiO₂ | K₂O | MgO | Na₂O | P₂O₅ |
|-------------|-----|------|-------|-------|-----|------|-----|-----|------|------|
| F1          | 2.99| 45.62| 35.66 | 3.38  | 1.16| 1.08 | 0.93| 0.50| 0.30 | 0.14 |
| F2          | 3.56| 50.24| 36.26 | 4.35  | 1.29| 0.98 | 0.97| 0.71| 0.23 |
| F3          | 5.17| 42.01| 35.87 | 4.71  | 2.63| 1.16 | 0.80| 0.81| 0.75 | 0.26 |

Tribromomethane (CHBr₃) and anhydrous ethanol (C₂H₆O) were used to prepare sorting solution with different densities, and their densities were 2.89 g/cm³ and 0.79 g/cm³ respectively.

The chemical reagents used to test pozzolanic activity of fly ash were solution of dilute hydrochloric acid and calcium hydroxide with concentrations of 15.7 g/L and 1.2 g/L, respectively.

2.2. Methods

Each original fly ash was divided into several particle groups according to bulk density by sorting liquid of water and solutions. The classified fly ash particles were used as research objects.

Pozzolanic activity of fly ash particles was measured by lime absorption method. The specific operation process was as follows: accurately weight 2g fly ash in a 100ml beaker, added 50ml calcium hydroxide solution, stirred evenly and then heated it in a water bath (65°C) for 24 hours. The beaker bottle should be sealed with plastic wrap so as to avoid the evaporation of water affecting the results of the experiment. Then the clear solution was extracted from the beaker with a 25ml pipette and placed in a 250 ml conical flask. 2 drops of methyl orange were added. The solution was titrated with solution of hydrochloric acid until the solution just turned red. The formula for calculating the fly ash pozzolanic activity index was as follows:

\[ \alpha = \frac{(0.06-0.043V \times 56)/2 \times 100}{ } \]  

where \( \alpha \) is pozzolanic activity index, \( V \) is the volume of hydrochloric acid solution consumed in liters.

The bulk density test method of graded fly ash particle group refers to the test process of cement density in GB/T 208-94. The liquid medium is water and anhydrous ethanol. The chemical composition of fly ash was determined by X-ray fluorescence spectrometer (XRF).

3 Results and discussions

3.1. Bulk density classification of original fly ash

Distilled water (1.00g/cm³), tribromomethane (2.89g/cm³) and anhydrous ethanol (0.79 g/cm³) were used for bulk density classification of original fly ash. The constituent of sorting liquids with different density were shown in table 2.

The mass percentage of fly ash particles in different bulk density ranges was shown in Figure 1. The bulk density distributions of three types of fly ash were significantly different, and the distribution of F2 was wider than that of F1 and F3. More than 98% of fly ash particles were located in the bulk density ranges of 1-2.89 g/cm³. The particles of bulk density less than 1 g/cm³ and greater than 2.89 g/cm³ were very few, and all particle contents of three types fly ash with bulk density greater than 2.89 g/cm³ were less than 0.05%. The particle contents with bulk density range of 2-2.5 g/cm³ were the highest, up to more than 70%.
Table 2. The constituent of sorting liquids wt%  

| Sorting liquid g/cm³ | Distilled water | Tribromomethane | Anhydrous ethanol |
|----------------------|-----------------|-----------------|-------------------|
| 1.00                 | 100             | 0.00            | 0.00              |
| 2.00                 | 0.00            | 57.63           | 42.37             |
| 2.50                 | 0.00            | 81.43           | 18.57             |
| 2.89                 | 0.00            | 100             | 0.00              |

Figure 1. The contents of fly ash particles in different bulk density ranges

3.2. Pozzonalic activity of fly ash particles with different bulk density

In the experiment, the pozzonalic activity of fly ash particles was tested by lime absorption method, and the results were shown in Figure 2.

According to the experimental results, the activity indexes of fly ash F1 and F2 decreased with the increase of bulk density, while that of fly ash F3 didn’t show the same change. In terms of overall activity, F1 and F3 were more active than F2.

Figure 2. Activity index of fly ash particles with different bulk density

In order to clarify the effect of different factors on the activity of fly ash particles, the relationships between activity and chemical composition or bulk density were analyzed with gray relationship analysis method.

Taking F1 as an example, the gray correlation coefficients of activity and bulk density or chemical
composition (Al$_2$O$_3$+SiO$_2$) were calculated. The Al$_2$O$_3$+SiO$_2$ content of three type fly ash was measured by XRF technology, and the results were shown in table 3.

### Table 3. Al$_2$O$_3$+SiO$_2$ content in different bulk density range of three type fly ash wt%

| Bulk density g/cm$^3$ | <1 | 1-2 | 2-2.5 | 2.5-2.89 |
|----------------------|----|-----|-------|---------|
| F1                   | 90.27 | 90.88 | 87.55 | 82.18 |
| F2                   | 88.82 | 87.14 | 86.12 | 83.74 |
| F3                   | 90.41 | 93.84 | 89.61 | 89.13 |

(1) Determine the analysis sequence $X_i(k)$, where the value of $i$ was 0,1,2, $k$ referred to the four bulk density segments from low to high as <1 g/cm$^3$, 1-2 g/cm$^3$, 2-2.5 g/cm$^3$, 2.5-2.89 g/cm$^3$, and the value was 1,2,3,4. Set X0 as the reference sequence, and the values were the activity values of fly ash particles in 4 bulk density ranges; set X1 and X2 as the comparison sequence, and the values were Al$_2$O$_3$+SiO$_2$ content and bulk density of fly ash particles in 4 bulk density ranges, as shown in table 4.

### Table 4. Grey relational analysis sequence of F1

| $k$ | 1 | 2 | 3 | 4 |
|-----|---|---|---|---|
| $X_0$ | 1.51 | 1.37 | 1.06 | 1.35 |
| $X_1$ | 90.27 | 90.88 | 87.55 | 82.18 |
| $X_2$ | 0.89 | 1.67 | 2.27 | 2.61 |

(2) Dimensionless $x_i(k) = \frac{X_i(k)}{X_i(1)}$. The results were shown in columns 2-4 of table 5.

(3) Calculate grey correlation coefficient $\xi$ by formula 2. The calculation process and results were shown in columns 5-8 of table 5.

$$\xi(k) = \frac{\min_i \min_k |x_i(k) - x_i(k)| + \rho \max_i \max_k |x_i(k) - x_i(k)|}{\max_i \max_k |x_i(k) - x_i(k)| + \rho \max_i \max_k |x_i(k) - x_i(k)|}$$  \hspace{1cm} (2)

Where, $\rho$ is the discrimination coefficient, and the value is 0.5.

### Table 5. Calculation process and result of grey correlation coefficient of F1

| $k$ | $x_0$ | $x_1$ | $x_2$ | $|x_0(k) - x_1(k)|$ | $|x_0(k) - x_2(k)|$ | $\xi_1(k)$ | $\xi_2(k)$ |
|-----|------|------|------|-------------------|-------------------|------------|------------|
| 1   | 1    | 1    | 1    | 0                 | 0                 | 1          | 1          |
| 2   | 0.9375 | 1.00676 | 1.8764 | 0.06926          | 0.9389           | 0.9474     | 0.57058    |
| 3   | 0.6875 | 0.96987 | 2.55056 | 0.28237          | 1.8631           | 0.81543    | 0.40106    |
| 4   | 0.4375 | 0.91038 | 2.93258 | 0.47288          | 2.4951           | 0.72514    | 0.3333    |

| min $|x_i - x_0|$ | 0 | 0 |
| max $|x_i - x_0|$ | 0.2824 | 2.4951 |

(4) Calculate correlation degree $r$, $r_i = \frac{1}{4} \sum_{k=1}^{4} \xi_i(k)$, $i=1,2$. It could be derived that $r_1(F1) = 0.8720$, $r_2(F1) = 0.5762$.

Correlation degrees of F2 and F3 were calculated according to the above process and it could be derived that $r_1(F2) = 0.9243$, $r_2(F2) = 0.5992$, $r_1(F3) = 0.9380$, $r_2(F3) = 0.5666$.

According to the above definition, $r_1$ and $r_2$ were the correlation degrees between activity with acid oxide content and bulk density respectively. The calculated results showed that all the correlation degrees between activity and acid oxide (Al$_2$O$_3$+SiO$_2$) content of three types fly ash were greater than 0.85, indicating that there was an obvious correlation between them. There was a positive correlation between activity and chemical composition, that was, the activity increased with the increase of acid oxide (Al$_2$O$_3$+SiO$_2$) content in fly ash. All correlation degrees between activity and bulk density of the three types fly ash were less than 0.6, indicating that there was no obvious correlation between them.
4. Conclusions

Original fly ash was classified by bulk density. The research object covered the whole range of the bulk density of fly ash particles, which provided theoretical data for practical sorting of fly ash. The bulk density of fly ash particles was concentrated in 1-2.5g/cm³, and the content of particles in this bulk density range exceeded 70%. The contents of particles with bulk density less than 1 g/cm³ and greater than 2.89 g/cm³ were very low, less than 2%. The bulk density distribution of different type fly ash particles was obviously different. The correlation degrees between activity and acid oxide content of the three types fly ash were 0.8720, 0.9243 and 0.9380, and the correlation degrees between activity and bulk density were 0.5762, 0.5992 and 0.5666. The results showed that there was an obvious positive correlation between acid oxide content and activity, while the effect of bulk density on activity was not obvious.

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