Study on particle size distribution of aggregate from coal ash for heat-resistant concrete

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Abstract. In Vietnam, the current amount of thermal ash (fly ash and bottom) remains nearly 100 million tons, causing great environmental and social impacts. The recycling and reuse of this source of waste is an effective solution towards sustainable development. The paper introduces the results of study on some properties and particle size distribution of aggregate from thermal ash, Duyen Hai factory for refractory concrete. Particle composition is calculated and selected according to the density of particle size arrangement with the maximum number of points of contact. The continuous particle size distributions of thermal ash is calculated by Andersen’s formula with Dₘₐₓ = 5 mm. The bulk density and porosity of the particle mixture corresponding to the vibrating modes is determined. By the experimental planning method (Design of experiments - DoE), the optimal aggregate particle composition was determined with the calculated value of n = 0.387 and the vibration time of 90 s gives the maximum bulk density of 1313.17 kg/m³ and the smallest actual porosity is 34.69%.

1. Introduction

Aggregates usually account for 50 ÷ 70% of the concrete's volume, so the properties of concrete at high temperatures are greatly influenced by the properties of the aggregate used. Natural aggregates such as sand, granite, gravel, limestone, etc. used for concrete production are only suitable at around 300-350°C. For quarried aggregates, due to the morphological changes of β-quartz causing the volume expansion, changes some physical properties of concrete at high temperature, weakens concrete, especially at 573°C. As for aggregates containing carbonate minerals, the carbonate dissociation at high temperature is one of the factors that destroy conventional concrete structures [2]. The aggregates used for reinforced concrete need to meet the requirements of thermal stability and high volumetric stability, not decomposing at high temperatures, preserving the structure of concrete under the effect of temperature. A number of studies have shown that to make aggregate for heat-resistant concrete, we can use materials durable at high temperatures (depending on the temperature of use) including natural minerals such as basalt, diabase, pumice stone and some artificial minerals such as chamotte [3-6], keramzite, fire-clay brick and tile wastes; bottom ash and slag metallurgy [7-9].

The properties of concrete usually depend not only on the type of aggregate used [9-10]. Heat-resistant concrete is not an exception to that rule. Most of the main properties of heat - concrete are greatly influenced not only by the type of aggregate, aggregate particle composition but also influenced by strength, thermal endurance, deformation temperature under load, fire resistance, volumetric stability of aggregate ... The particle size distributions is determined by content of each type of large aggregate size, small aggregate size, and the proportion between them. An aggregate with optimal particle size distributions when large particles act as a bearing frame, small particles play a compacting and filling
role to create a solid structure for heat-resistant concrete. Therefore, it is necessary to select and calculate the optimal particle size distributions [11]. According to Bozenov's principle [11], the selection of particle size distributions according to the density of grain size arrangement with the largest number of contact points plays an important role in the actual production of construction materials. In the production of concrete in general or heat resistant concrete in particular as well as in the production of ceramics, the method of calculation and selection of grain composition according to these principles is usually used. When the aggregate mixture has many particle grades, the ability to fill the gaps between the particles is easier. Therefore, It reduces the porosity, increases the contact area between particles, internal friction, stability, leading to increase the density and strength, volumetric stability, fire resistance, deformation temperature under the load of of concrete. In order to improve the ability of contact between particles, technology can be applied such as vibration method to increase the density of the aggregate mixtures.

In this paper, we introduce the method of designing the aggregate particle composition from ash and slag of the thermal power plant (D_max = 5 mm); Using the compacting technology modes with different vibration times to produce in the highest bulk density or lowest porosity of aggregate mixture. The actual porosity value was also determined for calculating the amount of water required for aggregate as well as the amount of binder used in the composition of concrete. The optimal particle composition of the ash aggregate is appropriately studied for making grout or small particle-resistant concrete.

2. Materials and experimental program

2.1. Materials

The material used in this work was coal ash (CA) from Duyen Hai thermal power plant (Vietnam). Chemical composition and physical properties is show in Table 1 and Table 2, respectively. The diffractograms of CA analyzed by XRD are shown in Fig. 1. The results indicate that the CA composes of quartz (SiO_2) and amorphous alumina silicate phase.

| Table 1. Chemical composition of coal ash (wt, %) |
| SiO_2 | Al_2O_3 | Fe_2O_3 | CaO | MgO | K_2O | Na_2O | SO_3 | L.O.I |
|-------|--------|--------|-----|-----|------|-------|------|------|
| 58.98 | 21.59  | 6.26   | 4.70| 2.27| 1.71 | 1.96  | 0.17 | 2.36 |

| Table 2. Properties of coal ash |
|------|------|------|------|
| N°   | Properties | Unit | Result |
| 1    | Density   | g/cm³ | 2.08  |
| 2    | Bulk density | kg/m³ | 1029  |
| 3    | Humidity  | %    | 0.27  |
| 4    | Water absorption | % | 11.87 |

When classifying the original slag grain sizes, according to the visual observations, the larger particle size has a rougher surface than the smaller particle size. The particles have a layered plate structure. Aggregate with particle size d > 5 mm have a flat rhomboid shape with many angles; with particle size d = 2.5 ÷ 5 mm, they are flat but more square; particle of size d < 2.5 mm has a more rounded shape, surface smoother than large particles. Particles with d > 5 mm when broken out have a crust layer structure, sometimes with small pores created by combustion of coal. In terms of color, slag particles mm are gray. The particle size d > 5 mm is relatively low (11.77%), a mechanical process is necessary for crushing particle of size d > 5 mm to size d ≤ 5 mm, then sieved to classify the particle sizes; fineness modulus of particles (Mdl) is to 1.95.
There have been many studies establishing formulas and charts to determine the proportion of particle sizes according to continuous or discontinuous particle size distribution [11-14]. In this paper, Andersen's formula for $D_{\text{max}} = 5$ mm was used as follows.

$$Y_i = \left(\frac{d_i}{D}\right)^n \times 100$$  \hspace{1cm} (1)

where: $Y_i$ is the content of particle sizes smaller than the given value $d_i$ (%);
$D$ is the largest particle size (mm);
$n$ is the empirically determined level index for each type of mixture and arrangement condition, $n = 0.33 - 0.5$.

A particle mixture achieves the optimal particle composition when the largest volume density or minimum porosity value is obtained. The actual porosity of the aggregate particle mixture including the porosity between aggregate particles and the voids is determined through the method of water volume occupying in the pore. This method is based on the amount of water introduced into the aggregate until the aggregate absorbs water to saturation, then calculates the amount of water occupying the hollow between aggregate particles, the amount of water absorbed into the voids in the particles; thereby calculating the porosity between particles, the porosity in the particles which in the theoretical formulas calculating the porosity cannot be determined. In order to increase the points of contact between particles, the aggregate mixture is mixed with the particle composition according to Andersen's formula, then compacted on the vibrating table with different vibration times such as 0s, 30s, 60s, 90s and 120s.

The particles mixture after vibration will be determined the bulk density, immersed in water until saturation state to determine the total actual porosity, the open porosity in the particles and the porosity between particles. In order to find the optimal particle composition that gives the largest bulk density of mixed particles, the paper used experimental planning method of Box and Hunter [15].

3. Results and discussion

3.1. Calculation of particle size distribution according to Andersen formula

The composition of thermal slag aggregate particles is calculated based on Andersen's formula with $D_{\text{max}} = 5$ mm, the value of $n = 0.33 \div 0.5$ is shown in Table 3:
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density and porosity of thermal ash aggregate


Thus, if the index (n) increases, the content of the coarse particle size increases, causing
the finest modulus Mdl of particle to increase significantly. Therefore, when calculating the
continuous grain composition according to Andersen formula with large (n) value, there will be a
crude grain mixture, meaning the porosity of the aggregate mixture increases. Therefore, it is
necessary to change the value of (n) to determine the optimal particle composition with appropriate
vibration technology mode to achieve the most compacted arrangement, the highest density, the lowest
porosity. Thus, through a factor of value (n), the content of particle sizes (rough, fine) from 5 to < 0.14
mm can be determined to achieve the highest value of bulk density; This method is similar to that of
other authors [11], [12] and [14].

3.2. The density and porosity of the particle mixture corresponds to the vibration technology

The volumetric density and porosity of thermal ash aggregate are calculated according to Andersen
formula (n = 0.33 ± 0.5) corresponding to the different compacting technology regimes (vibrating for
0 s, 30 s, 60 s, 90 s, 120 s) are presented in Table 4. In the natural bulk state (without vibrating - 0s),
when the index n increases from 0.33 ± 0.50, the density of the particle mixture decreases and the
porosity increases due to the increase of the large particle content and the reduction of the small
particle content. It can be explained by the fact that the large particles themselves have porous hollow
structures, small volume density, the small particles will not be enough to fill the gap between large
particles. When the compacting mode of aggregate mixture is applied, the value of the volume density
increases. When the vibrating time increases from 0 s to 90 s, the value of the volumetric density of
the aggregate particle mixture increases gradually and if the compaction time increases > 90 s, the
value of the volume density of the aggregate particle mixture tends to decrease due to already reaching
an optimal level of compaction, vibration will cause loose the particle mixture. In the vibrating mode
of 90 s, when n = 0.33 ± 0.4, the volume increases and the porosity decreases, but when n > 0.4, the

| Index n | Yi, ai, Ai (%) | Sieve (mm) | M_{dl} |
|---------|----------------|------------|--------|
| Yi      | 100            | 2.5        | 1.25   | 0.63   | 0.315 | 0.14 | < 0.14 |
| ai      | 20.45          | 63.29      | 50.48  | 40.16  | 30.73 |
| Ai      | 20.45          | 36.71      | 49.52  | 59.84  | 69.27 |
| Yi      | 100            | 37.38      | 59.87  | 46.47  | 35.95 |
| ai      | 22.62          | 17.5       | 13.41  | 10.51  | 9.32  |
| Ai      | 22.62          | 40.13      | 53.53  | 64.05  | 73.37 |
| Yi      | 100            | 75.79      | 57.43  | 43.67  | 33.09 |
| ai      | 24.21          | 18.36      | 13.76  | 10.58  | 9.16  |
| Ai      | 24.21          | 42.57      | 56.33  | 66.91  | 76.07 |
| Yi      | 100            | 74.23      | 55.1   | 41.04  | 30.46 |
| ai      | 25.77          | 19.13      | 14.04  | 10.58  | 8.97  |
| Ai      | 25.77          | 44.9       | 58.96  | 69.54  | 78.51 |
| Yi      | 100            | 73.20      | 53.59  | 39.37  | 28.82 |
| ai      | 26.80          | 19.62      | 14.22  | 10.55  | 8.81  |
| Ai      | 26.80          | 46.41      | 60.63  | 71.18  | 79.99 |

When the value of n varies from 0.33 to 0.5, the amount of different particle sizes varies depending the
particle size. With the particle size of 2.5 ÷ 5 mm, 1.25 ÷ 2.5 mm, 0.63 ÷ 1.25 mm, an increase of
8.84%, 4.45%, 1.69% respectively is remarked; With the particle size of mm, the increase is negligible;
But with particle size 0.14 ÷ 0.315 mm and < 0.14mm, the reduction of the content of these particle is
1.06% - 14%. Thus, if the index (n) increases, the content of the coarse particle size increases, causing
the finest modulus Mdl of particle to increase significantly. Therefore, when calculating the
continuous grain composition according to Andersen formula with large (n) value, there will be a
crude grain mixture, meaning the porosity of the aggregate mixture increases. Therefore, it is
necessary to change the value of (n) to determine the optimal particle composition with appropriate
vibration technology mode to achieve the most compacted arrangement, the highest density, the lowest
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| Ai      | 26.80          | 46.41      | 60.63  | 71.18  | 79.99 |
| Yi      | 100            | 70.71      | 50.00  | 35.5   | 25.1  |
| ai      | 29.29          | 20.71      | 14.5   | 10.4   | 8.37  |
| Ai      | 29.29          | 50.00      | 64.5   | 74.9   | 83.27 |
volume density decreases and the porosity increases. At the value of \( n = 0.40 \), the number of small particles decreases sufficiently to fill between large particles, so the volume density reaches the maximum value, the smallest porosity.

Table 4. Bulk density, porosity of aggregate mixtures with different time

| n   | Vibration time \( t \) (s) | Volume density \( \gamma_d \) (kg/m\(^3\)) | \( r_1 \) (%) | \( r_2 \) (%) | Porosity, \( r \) (%) | Difference \( r_1 - r_2 \) (%) |
|-----|--------------------------|----------------------------------------|--------------|--------------|----------------------|--------------------------|
| 0.33| 0                        | 1115.3                                 | 46.46        | -            | -                    | -                        |
|     | 30                       | 1275.0                                 | 38.79        | 35.87        | 4.42                 | 31.45                    |
|     | 60                       | 1295.1                                 | 37.83        | 35.26        | 4.61                 | 30.65                    |
|     | 90                       | 1272.9                                 | 38.89        | 36.12        | 4.57                 | 31.55                    |
|     | 120                      | 1248.7                                 | 40.05        | 37.87        | 4.11                 | 33.76                    |
| 0.37| 0                        | 1088                                   | 47.77        | -            | -                    | -                        |
|     | 30                       | 1251.9                                 | 39.90        | 36.94        | 4.58                 | 32.36                    |
|     | 60                       | 1279.2                                 | 38.59        | 35.91        | 4.79                 | 31.12                    |
|     | 90                       | 1294.9                                 | 37.83        | 35.56        | 4.89                 | 30.67                    |
|     | 120                      | 1260.1                                 | 39.51        | 37.29        | 4.60                 | 32.69                    |
| 0.4 | 0                        | 1072                                   | 48.54        | -            | -                    | -                        |
|     | 30                       | 1236.9                                 | 40.62        | 37.79        | 4.65                 | 33.14                    |
|     | 60                       | 1276.2                                 | 38.73        | 36.21        | 4.83                 | 31.38                    |
|     | 90                       | 1305.9                                 | 37.31        | 34.96        | 4.94                 | 30.02                    |
|     | 120                      | 1268.1                                 | 39.12        | 36.23        | 4.79                 | 31.44                    |
| 0.43| 0                        | 1060.6                                 | 49.08        | -            | -                    | -                        |
|     | 30                       | 1222.2                                 | 41.33        | 38.50        | 4.71                 | 33.79                    |
|     | 60                       | 1264.9                                 | 39.28        | 36.80        | 4.92                 | 31.88                    |
|     | 90                       | 1282.7                                 | 38.42        | 35.85        | 5.02                 | 30.83                    |
|     | 120                      | 1242.6                                 | 40.35        | 38.04        | 4.84                 | 33.20                    |
| 0.45| 0                        | 1043.4                                 | 49.91        | -            | -                    | -                        |
|     | 30                       | 1215.6                                 | 41.64        | 39.01        | 4.95                 | 34.06                    |
|     | 60                       | 1252.7                                 | 39.86        | 37.30        | 5.12                 | 32.18                    |
|     | 90                       | 1273.9                                 | 38.84        | 36.45        | 5.34                 | 31.11                    |
|     | 120                      | 1228.9                                 | 41.00        | 38.67        | 5.09                 | 33.58                    |
| 0.5 | 0                        | 1006.9                                 | 51.66        | -            | -                    | -                        |
|     | 30                       | 1179.6                                 | 43.37        | 40.54        | 5.17                 | 35.37                    |
|     | 60                       | 1230.4                                 | 40.93        | 38.54        | 5.39                 | 33.15                    |
|     | 90                       | 1247.5                                 | 40.11        | 37.94        | 5.50                 | 32.44                    |
|     | 120                      | 1232.8                                 | 40.82        | 38.00        | 5.44                 | 32.56                    |

where: \( r_1^{tt} \) is the theoretical total porosity (%); 
\( r_2^{tt} \): is the actual porosity determined by the method of absolute water volume (%); 
\( r_1 \) is the open porosity in aggregate particle determined by the method of absolute water volume (%);
r₂ is the porosity between aggregate particle determined by the method of absolute water volume (%). When comparing the values of porosity, the results shows that r₂ is greater than r₁. The reason of this result is that the total porosity of aggregates when calculating in theory is included closed voids; But when calculating the actual volume based on water occupying in open pores of aggregate, the water cannot penetrate into the aggregate closed void, especially the pore size smaller than 0.1µm. Through the actual porosity, the amount of water mixed with mortar and concrete will be determined more accurately, avoiding excess water affecting the heat resistance and thermal endurance of mortar and concrete.

3.3. Optimal particle size distribution

In order to determine the optimal particle size distribution of thermal ash aggregates mixtures with Dₘₐₓ = 5 mm corresponding to the 90 s vibration mode, the article has set up an experimental planning model, solving to find the optimal function of the aggregate mixture with the influence factor is the index (n), the objective function is the value of the volume density, the actual porosity with the value at the planning center is n = 0.43 and the interval plan of 0.02; The experimental planning problem applies Box and Hunter’s quadratic center plan with an influencing factor, the value of the step is determined by 2¹/₄ [15]. The coding variables and experimental planning matrix of aggregate distribution are presented in Tables 5 and 6.

| Actual variables | Coding variables | Planning levels | Jump in planning |
|------------------|------------------|-----------------|-----------------|
| n, x             | -2¹/₄           | -1             | 0               | 0.02            |
|                 | 0.376           | 0.38           | 0.40            | 0.42            |
|                 | 0.424           |                |                 |                 |

Table 5. Coding variables in experimental planning of particle composition.

| No  | Coding variables | Volume density of aggregate mixture (kg/m³) | Actual porosity of the aggregate mixture (%) |
|-----|------------------|---------------------------------------------|---------------------------------------------|
| 1   | +1               | 1287.6                                     | 35.45                                       |
| 2   | -1               | 1328.2                                     | 34.33                                       |
| 3   | +2¹/₄            | 1285.3                                     | 35.67                                       |
| 4   | -2¹/₄            | 1298.8                                     | 35.09                                       |
| 5   | 0                | 1305.9                                     | 34.99                                       |
| 6   | 0                | 1311.1                                     | 34.70                                       |
| 7   | 0                | 1318.3                                     | 34.65                                       |
| 8   | 0                | 1302.1                                     | 34.99                                       |
| 9   | 0                | 1304.1                                     | 34.84                                       |

where: Yᵢ, rᵢ is the value of the volume, the porosity of the sample i (i = 1, 2, 3) (kg/m³);
Y̅ᵢ, r̅ is the average volumetric density (kg/m³), average porosity (%) of samples.

Solving experimental planning problem with maple software 13.0, properties of the mixture of ash aggregate slag with Dₘₐₓ = 5 mm have the following important results:

+ The regression function for volume density is as follows: 
  \[ \bar{Y}_{\text{r}} = 1309.3396 - 11.5409x - 8.7119x^2 \],

+ The regression function for porosity is as follows: 
  \[ \bar{r} = 34.80 + 0.3749x + 0.3011x^2 \].
The maximum value of the volumetric density of aggregate mixture is \( Y = 1313.17 \text{ (kg/m}^3) \) at \( x=0.6627 \) or \( n = 0.387 \). The actual minimum value of the porosity of the mixture of thermal aggregate is \( Y = 34.69\% \) at \( x = 0.6258 \) or \( n = 0.387 \). Thus, the aggregate from the thermal power plant has the optimal particle composition with a value of \( n = 0.387 \) corresponding to the vibration time of 90 s.

4. Conclusions

Based on the empirical results, some conclusions are drawn as follows:

- Using Andersen's formula and vibration process to find the optimum particle size distribution which give the highest volume density and porosity.
- With the method of absolute water volume occupied in the pore, the values of open porosity and porosity between particle have been determined. This give the direction for determining the amount of water needed for the aggregate mixture from which the suitable amount of binder in heat resistant concrete.
- By the experimental planning method, the optimal particle composition of Duyen Hai thermal ash and slag aggregate was found suitable for manufacturing concrete with the index of \( n = 0.387 \) and the compaction time is 90 s to achieve the maximum mass density of 1313.17 kg/m\(^3\) and the smallest porosity is 34.69\%.

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