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A new method for evaluating packing efficiency of particle size distribution during coal water slurry preparation

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Abstract: Particle size distribution is a key problem in preparation of coal water slurry . Due to the large number of particles and irregular accumulation of particles, how to evaluate and calculate CWS particles has become an urgent need. In this paper, the volume accumulation of particles is simplified as plane accumulation, and the particle packing gradation model based on the three-particle packing gradation and the four-particle packing gradation is constructed. Based on the classical mathematical principle, the packing voids and packing probability of 5 typical CWS particle sizes were calculated. The packing styles of $N_1N_2N_3$, $N_2N_3N_4$ and $N_3N_4N_5$ were evaluated and analyzed. The evaluation method of packing index based on $\Sigma P \times S_{\min}$ and $\Sigma P \times S_{\max}$ was established. The results show that the packing efficiency of particles can be quantitatively evaluated by using of cumulative calculation of voids areas and packing probabilities under different packing modes. The evaluation results are consistent to the tendency of CWS concentration. By this method, the particle size distribution of CWS can be controlled and adjusted during both wet milling processes and experimental investigations. Furthermore, the method may be used to other particle packing cases.

Key words: probability analysis; packing efficiency; void distribution; particle size distribution; coal water slurry

Introduction

The packing property of particles refers to the inner part of particles, the arrangement of particles in space or the structural characteristics of particles. The preparation of high concentration of coal water slurry requires higher packing density. In the study of dense packing of particles, there are two major theories: continuous and discontinuous packing of particles. Continuously distributed particles are composed of particles of all sizes within a certain range of particle size, the discontinuous distribution is composed of particles representing the finite size of the range. Studies on accumulation of discrete-size particles. (Furnas 1928; Westman et al 1930; Westman 1936; Suzuki 1985) The initiator of the classical continuous theory was Andreason (Andreason 1928; Andreason 1929; Andreason 1930), who described the actual particle distribution with equation of Gaudin-Schuhmann Golden-schutzmann Fuller (Fuller 1906) obtained the experience curve of the most compact packing of continuous grain-size system by experiment. It is considered that the equation modulus m in the range of 0.33 ~ 0.5 has the minimum void fraction. Based on Andreason's theory, the Alfred equation is proposed by Dinger and Funk (Funk 1980; Funk 1987), and it is found that the highest accumulation rate occurs when n is 0.37. Some researchers put forward the theory of inter-stage packing by analytic method, and established a new relation between the modulus n and the void fraction of Gaudin and Alfred equation, and calculated the packing efficiency of any particle size distribution (Zhang et al 2002).

With the rapid development of China's coal chemical industry, the coal production process of methanol, synthetic ammonia, olefins by using of coal water slurry as gasification media is expected to use 300 million tons in the future, the amount of CWS used for combustion is about 30-60 million tons annually. Therefore, coal-water slurry gasification has become the focus of chemical industry and the preparation of high concentration coal-water slurry has become an important factor in CWS applications. The minimum void accumulation model and evaluation index E provides a good method to testify the coarse particle and fine particle distribution (Tu 2013; Tu et al 2013; Tu et al 2015; Yang et al 2016). Increasing the size range of coal water slurry particles is beneficial to improving its packing efficiency, the packing efficiency is related to the particle size composition of raw materials (Chen et al 2012). (Huang et al 2006) provide a formula for calculating the packing density of powder with continuous particle size in slurry. (Guo et al 2006; Chen 2016; Zhou 2001) put forward the discontinuous grading model and emphasize the function of the coarse particles and fine particles. (Yang et al 2015) describe a multi-peak gradation to evaluate the particle size distribution of lignite slurry after grinding with fractal theory. (Huang et al 1986) used single peak model to testify the particle size distribution with low accumulated voids. (Zhang et al 2004; Jiao 2007) have established three creative models of particle number distribution model, particle surface area distribution model and particle mass distribution model to describe the dynamic change of the particle size distribution during coal grinding. (Wang et al 2013; Wang et al 2014) calculate the particle size of Shenfu coal powder by Alfred Model.

However, sometimes it is difficult for engineers in CWS factories to understand these formulas. A

simplified description and method for packing efficiency based on particle size distribution of CWS is needed.

2. Experimental methods and procedures

Fig.1 is the experimental diagram for particles evaluation. The size composition of CWS consists of 8-mesh, 14-mesh, 40-mesh, 80-mesh and 200-mesh. Most of the size composition of qualified CWS from 8 to 14-mesh is less than 1% . Therefore, five particle sizes of 14-40 mesh, 40-80 mesh, 80-200 mesh, 200-325 mesh, <325 Mesh were selected as the research objects. According to the packing model in Fig. 2, the packing voids of particles in CWS can be calculated simply by the distribution of the above five kinds of particle sizes.

Figure 2 is the model of particle packing gradation. In Figure 2, $R_1R_1R_1$, $R_1R_1R_2$, $R_1R_2R_2$, $R_2R_2R_2$, $R_1R_1R_3$, $R_1R_2R_3$, $R_1R_3R_3$, $R_2R_3R_3$ and $R_3R_3R_3$ are packed in a steady state with a radius of R_1 , R_2 , R_3 . $R_1R_1R_1R_1$, $R_1R_1R_1R_2$, $R_1R_1R_2R_2$, $R_1R_2R_1R_2$, $R_1R_1R_1R_3$, $R_2R_2R_2R_1$, $R_1R_1R_2R_3$, $R_1R_2R_1R_3$, $R_2R_2R_2R_2$, $R_1R_2R_2R_3$, $R_1R_2R_3R_2$, $R_1R_1R_3R_3$, $R_1R_3R_1R_3$, $R_2R_2R_2R_3$, $R_1R_2R_3R_3$, $R_1R_3R_2R_3$, $R_2R_2R_3R_3$, $R_2R_3R_2R_3$, $R_3R_3R_3R_1$, $R_3R_3R_3R_2$, $R_3R_3R_3R_3$ are packed in a non-steady state with a radius of R_1 , R_2 , R_3 .

The packing voids of particles in a unit volume is calculated and analyzed by the voids formed by particles in its cross section. The calculation procedure is as follows:

1) the particle volumes of the above five regions are defined as V_1 , V_2 , V_3 , V_4 , V_5 . The mass of a single particle in the five size range is defined as M_1 , M_2 , M_3 , M_4 , and M_5 . The number of particles per unit mass in the five size ranges is defined as W_1 , W_2 , W_3 , W_4 , W_5 . The number of particles per unit mass cross section is defined as N_1 , N_2 , N_3 , N_4 , N_5 for each of the five dimensions.

2) the probability distribution, void area distribution and cumulative void area of 10 particle grading modes under $N_1N_2N_3$ and $N_3N_4N_5$ packing modes were calculated according to the three-particle steady state mechanism of graded packing.

3) the probability distribution, void area distribution and cumulative void area of 21 kinds of particle grading modes under $N_1N_2N_3$ and $N_3N_4N_5$ packing modes are calculated according to the four-particle unsteady mechanism of graded packing.

The median of the granularity range is used as the value of the granularity range. The values of D_1 , D_2 , D_3 , D_4 and D_5 are $940\mu\text{m}$, $325\mu\text{m}$, $138\mu\text{m}$, $60\mu\text{m}$ and $22\mu\text{m}$ respectively. As coal particles are non-spherical particles, the spherical degree of broken particles is 0.75, so the diameter of the non-spherical particles is about 0.75 times diameter of the spherical particles. The particle diameters of the samples were $705\mu\text{m}$, $244\mu\text{m}$, $104\mu\text{m}$, $45\mu\text{m}$ and $17\mu\text{m}$ respectively. The calculation is as follows:

$$\begin{aligned} D_1 &= \frac{(14\text{mesh} + 40\text{mesh})}{2} = \frac{(1430\mu\text{m} + 450\mu\text{m})}{2} = 940\mu\text{m} & D_1' &= 0.75D_1 = 0.75 \times 940\mu\text{m} = 705\mu\text{m} \\ D_2 &= \frac{(40\text{mesh} + 80\text{mesh})}{2} = \frac{(450\mu\text{m} + 200\mu\text{m})}{2} = 325\mu\text{m} & D_2' &= 0.75D_2 = 0.75 \times 325\mu\text{m} = 243.75\mu\text{m} \approx 244\mu\text{m} \\ D_3 &= \frac{(80\text{mesh} + 200\text{mesh})}{2} = \frac{(200\mu\text{m} + 76\mu\text{m})}{2} = 138\mu\text{m} & D_3' &= 0.75D_3 = 0.75 \times 138\mu\text{m} = 103.5\mu\text{m} \approx 104\mu\text{m} \\ D_4 &= \frac{200\text{mesh} + 325\text{mesh}}{2} = \frac{76\mu\text{m} + 44\mu\text{m}}{2} = 60\mu\text{m} & D_4' &= 0.75D_4 = 0.75 \times 60\mu\text{m} = 45\mu\text{m} \\ D_5 &= \frac{325\text{mesh} + 0}{2} = \frac{44\mu\text{m}}{2} = 22\mu\text{m} & D_5' &= 0.75D_5 = 0.75 \times 22\mu\text{m} = 16.5\mu\text{m} \approx 17\mu\text{m} \end{aligned}$$

For the purposes of the study, the particle is approximately considered to be a sphere with a volume of 1mm^3 . The density of coal particles is 1.35g/cm^3 . The volume and mass of the coal particles in each size range are calculated as follows:

$$\begin{aligned}
V_1 &= \frac{\pi D_1^3}{6} = 1.83 \times 10^{-10} \text{ m}^3, \quad m_1 = V_1 \times \rho = 2.47 \times 10^{-4} \text{ g}; \\
V_2 &= \frac{\pi D_2^3}{6} = 7.58 \times 10^{-12} \text{ m}^3, \quad m_2 = V_2 \times \rho = 1.02 \times 10^{-5} \text{ g}; \\
V_3 &= \frac{\pi D_3^3}{6} = 5.81 \times 10^{-13} \text{ m}^3, \quad m_3 = V_3 \times \rho = 7.84 \times 10^{-7} \text{ g}; \\
V_4 &= \frac{\pi D_4^3}{6} = 4.77 \times 10^{-14} \text{ m}^3, \quad m_4 = V_4 \times \rho = 6.44 \times 10^{-8} \text{ g}; \\
V_5 &= \frac{\pi D_5^3}{6} = 2.35 \times 10^{-15} \text{ m}^3, \quad m_5 = V_5 \times \rho = 3.17 \times 10^{-9} \text{ g};
\end{aligned}$$

The particle size distribution of 4 CWS samples are listed in Table 1 .

Samples A, B and C are coal water slurry samples from a methanol plant in Ningxia, and Samples D are coal water slurry samples from a methanol plant in Inner Mongolia. The particle size of each group was the average value of 10 CWS samples. The particle distribution parameters of samples A, B, C and D are calculated in Table 2.

3.1 Average size

According to the particle size distribution in Table 1, the average particle size values of A, B, C and D CWS samples are 170 μm , 175 μm , 170 μm and 186 μm respectively. The calculation is as follows:

$$\bar{D} = \sum_{i=0}^n D_i W_i = D_1 W_1 + D_2 W_2 + D_3 W_3 + D_4 W_4 + D_5 W_5 \quad \text{Formula (1)}$$

Comparing the concentration values of A, B, C and D samples, it is found that the concentration values of D samples are 55.07% , and the average size values of slurry are 186 μm , and the average size difference is larger than that of A, B and C. The concentration difference of A, B and C samples was 1-2wt% , and the average particle size difference was 5 μm . It is concluded that the larger the average size of CWS, the larger the number of coarse particles in CWS. The reason for this result lies in the incomplete grinding of CWS particles by steel rods in rod mills during wet overflow milling, causing more coarse particles to overflow into the finished slurry. The 14-40 mesh particle ratio of D sample in Table 1 is obviously higher than that of A, B and C sample.

3.2 Three-particle packing gradation

3.2.1 Heron Formula

The size of the void formed between particles can be expressed by the area of the triangle formed by the three central points of particles. As shown in Figure 3(a) , (b) in the triangle ABC. The area of the triangle can be calculated by Heron's formula (Xia 2007) . See Formula 2.

$$\begin{aligned}
S_{\min} &= \sqrt{s(s-a)(s-b)(s-c)} \\
s &= \frac{(a+b+c)}{2}
\end{aligned} \quad \text{Formula (2)}$$

There are 10 packing modes according to the three-particle packing gradation, which are $R_1 R_1 R_1, R_1 R_1 R_2, R_1 R_2 R_2, R_2 R_2 R_2, R_1 R_1 R_3, R_1 R_2 R_3, R_2 R_2 R_3, R_1 R_3 R_3, R_2 R_3 R_3$ and $R_3 R_3 R_3$, respectively.

According to the Formula (2) , the calculation values of the void areas of the three-particle

packing in $N_1N_2N_3$ and $N_3N_4N_5$ packing modes are shown in Table 3.

Meanwhile, the probability of $R_1R_1R_1, R_1R_1R_2, R_1R_2R_2, R_2R_2R_2, R_1R_1R_3, R_1R_2R_3, R_2R_2R_3, R_1R_3R_3, R_2R_3R_3$ and $R_3R_3R_3$ packing modes can be calculated according to the formula (3) .

$$\begin{aligned}
 P(R_1R_1R_1) &= \frac{C_{N_1}^3}{C_N^3}; \quad P(R_1R_1R_2) = \frac{C_{N_1}^2 \times C_{N_2}^1}{C_N^3}; \quad P(R_1R_2R_2) = \frac{C_{N_1}^1 \times C_{N_2}^2}{C_N^3}; \\
 P(R_2R_2R_2) &= \frac{C_{N_2}^3}{C_N^3}; \quad P(R_1R_1R_3) = \frac{C_{N_1}^2 \times C_{N_3}^1}{C_N^3}; \quad P(R_1R_2R_3) = \frac{C_{N_1}^1 \times C_{N_2}^1 \times C_{N_3}^1}{C_N^3}; \\
 P(R_2R_2R_3) &= \frac{C_{N_2}^2 \times C_{N_3}^1}{C_N^3}; \quad P(R_1R_3R_3) = \frac{C_{N_1}^1 \times C_{N_3}^2}{C_N^3}; \quad P(R_2R_3R_3) = \frac{C_{N_2}^1 \times C_{N_3}^2}{C_N^3}; \quad P(R_3R_3R_3) = \frac{C_{N_3}^3}{C_N^3};
 \end{aligned}
 \tag{3}$$

In formula (3) ,

$$\begin{aligned}
 &P(R_1R_1R_1) + P(R_1R_1R_2) + P(R_1R_2R_2) + P(R_2R_2R_2) + P(R_1R_1R_3) + \\
 &P(R_1R_2R_3) + P(R_2R_2R_3) + P(R_1R_3R_3) + P(R_2R_3R_3) + P(R_3R_3R_3) = 1
 \end{aligned}$$

3.2.2 Probability distribution of three-particle packing gradation

According to the Formula (3) , the probability distribution value P of each packing mode can be calculated. The probability distribution of coal water slurry samples A, B, C and D is shown in Fig.4.

In Fig. 4, $N_1N_2N_3$ is the packing probability distribution of particles with diameter D_1, D_2, D_3 and number of particles N_1, N_2, N_3 . $N_3N_4N_5$ is the packing probability distribution of particles with diameter D_3, D_4, D_5 and number of particles N_3, N_4, N_5 . As can be seen from the diagram, the probability of three particles forming $R_3R_3R_3$ is the highest in the $N_1N_2N_3$ grading model, which is 66.4% , 65.3% ,65.2% and 64.2% respectively, followed by $R_2R_3R_3$ packing. Under the $N_2N_3N_4$ packing gradation mode, the probability of three particles forming $R_2R_3R_3$ packing is the highest which are 38.4%, 37.9%, 38.7%, 38.5% respectively, followed by $R_3R_3R_3$ and $R_2R_2R_3$. Under the $N_3N_4N_5$ packing gradation mode, the probability of three particles forming $R_3R_3R_3$ packing is the highest which is 84.7% , 84.6%, 82.8% and 81.9% respectively, followed by $R_1R_3R_3$ and $R_2R_3R_3$. The probabilities of packing modes of $R_1R_1R_1, R_1R_1R_2, R_1R_2R_2, R_2R_2R_2, R_1R_1R_3, R_1R_2R_3, R_2R_2R_3$ are small and their accumulative probabilities are 0.8%, 0.9%, 1.1% and 1.2% respectively.

3.2.3 Distribution of void areas in three-particle packing gradation

According to the calculation value S_{min} in Formula (2) and the probability value P in formula (3) , the value of $P \times S_{min}$ in each packing mode can be calculated. The probability distribution of three-particle packing model and the distribution of packing void areas of CWS samples A, B, C and D are shown in Fig. 5.

As can be seen from the Fig.5, in the $N_1N_2N_3$ packing gradation mode, the contribution of $R_3R_3R_3$ packing of three particles to the void area is the largest, which are $3107\mu m^2, 3059\mu m^2, 3053\mu m^2, 3005\mu m^2$ respectively, followed by $R_2R_3R_3$. Under the $N_2N_3N_4$ packing gradation mode, the contribution of $R_2R_3R_3$ to the packing void area is the largest, which are $613\mu m^2, 606\mu m^2, 619\mu m^2$ and $616\mu m^2$ respectively, followed by $R_2R_2R_3, R_1R_2R_3$ and $R_3R_3R_3$. And the contribution of different CWS samples to the packing void area varies alternately. Under the $N_3N_4N_5$ packing gradation mode, the contribution of $R_3R_3R_3$ to the packing void area is the largest, which are $106\mu m^2, 106\mu m^2, 104\mu m^2$ and $102\mu m^2$ respectively, followed by $R_2R_3R_3$ and $R_1R_3R_3$. The probabilities of packing void area of $R_1R_1R_1, R_1R_1R_2, R_1R_2R_2, R_2R_2R_2, R_1R_1R_3, R_1R_2R_3, R_2R_2R_3$ are small and their accumulative probabilities are $7.0\mu m^2, 7.2\mu m^2, 8.5\mu m^2$ and $9.0\mu m^2$ respectively.

3.2.4 Cumulative void area of three-particle packing gradation

Based on the calculation of the probability distribution of packing gradation and the area distribution, the cumulative void area of packing gradation $\sum P \times S_{\min}$ is calculated in order to further analyze the difference of packing void in different granular packing states, see Formula (4).

$$\begin{aligned} \sum P \times S_{\min} = & P(R_1 R_1 R_1) \times S_{\min}(R_1 R_1 R_1) + \\ & P(R_1 R_1 R_2) \times S_{\min}(R_1 R_1 R_2) + P(R_1 R_2 R_2) \times S_{\min}(R_1 R_2 R_2) + \\ & (R_2 R_2 R_2) \times S_{\min}(R_2 R_2 R_2) + P(R_1 R_1 R_3) \times S_{\min}(R_1 R_1 R_3) + P(R_1 R_2 R_3) \times S_{\min}(R_1 R_2 R_3) + \\ & P(R_2 R_2 R_3) \times S_{\min}(R_2 R_2 R_3) + P(R_1 R_3 R_3) \times S_{\min}(R_1 R_3 R_3) + P(R_2 R_3 R_3) \times S_{\min}(R_2 R_3 R_3) + P(R_3 R_3 R_3) \times S_{\min}(R_3 R_3 R_3); \end{aligned}$$

Formula (4)

The cumulative packing void areas of CWS samples A, B, C and D is shown in Fig.6.

It can be seen from Fig.6 that under the condition of $N_1 N_2 N_3$ packing gradation, the accumulative packing void areas of samples A, B, C and D are $6626 \mu m^2$, $6694 \mu m^2$, $6695 \mu m^2$ and $6865 \mu m^2$ respectively. The accumulated accumulation void area of D sample is the highest. Under the condition of $N_2 N_3 N_4$ packing gradation, the accumulative packing voids area values of samples A, B, C and D are $2275 \mu m^2$, $2338 \mu m^2$, $2130 \mu m^2$, $1957 \mu m^2$ respectively. The accumulated void area of D sample is the lowest. This result is contrary to the conclusion that the higher the packing efficiency the higher the slurry concentration. The results are consistent with the conclusion that the higher the packing efficiency the higher the CWS concentration. The reason is that $N_2 N_3 N_4$ is a concentrated packing with median particle size. According to Alfred Model, the most compact packing can be achieved when fine coal powder can be completely filled into coarse coal powder. Due to the small packing voids of medium-sized pulverized coal, fine pulverized coal can not fill the voids formed by medium-sized pulverized coal, which results in the decrease of packing voids and the decrease of slurry concentration. Under the condition of $N_3 N_4 N_5$ packing gradation, the accumulative packing voids area values of samples A, B, C and D are $169 \mu m^2$, $170 \mu m^2$, $172 \mu m^2$, $172 \mu m^2$, respectively. The results are consistent with the conclusion that the higher the packing efficiency the higher the CWS concentration. In conclusion, it is feasible to use cumulative probability gap area $\sum P \times S_{\min}$ to evaluate the packing efficiency of CWS.

3.3 Four-particles packing gradation

3.3.1 Bretschneider Formula

The size of the voids between the particles can be expressed by the area of the quadrilateral which is connected by the four central points of the particles. As shows in Figures 2(c) and (d), the quadrangle ABCD. The area of the quadrilateral can be calculated using Bretschneider formula (Shan 2002). See Formula 5.

$$S_{\max} = \sqrt{(p-a)(p-b)(p-c)(p-d)}$$

$$p = \frac{(a+b+c+d)}{2}$$

Formula (5)

According to the four particle packing gradation, there are 21 packing gradation modes, which are $R_1 R_1 R_1 R_1$, $R_1 R_1 R_1 R_2$, $R_1 R_1 R_2 R_2$, $R_1 R_2 R_1 R_2$, $R_1 R_1 R_1 R_3$, $R_2 R_2 R_2 R_1$, $R_1 R_1 R_2 R_3$, $R_1 R_2 R_1 R_3$, $R_2 R_2 R_2 R_2$, $R_1 R_2 R_2 R_3$, $R_1 R_2 R_3 R_2$, $R_1 R_1 R_3 R_3$, $R_1 R_3 R_1 R_3$, $R_2 R_2 R_2 R_3$, $R_1 R_2 R_3 R_3$, $R_1 R_3 R_2 R_3$, $R_2 R_2 R_3 R_3$, $R_2 R_3 R_2 R_3$, $R_3 R_3 R_3 R_1$, $R_3 R_3 R_3 R_2$ and $R_3 R_3 R_3 R_3$.

According to the Formula (5), the void areas of four-particle packing in $N_1 N_2 N_3$ and $N_3 N_4 N_5$

packing modes are shown in Table 4.

Meanwhile, the probability of $R_1R_1R_1R_1$, $R_1R_1R_1R_2$, $R_1R_1R_2R_2$, $R_1R_2R_1R_2$, $R_1R_1R_1R_3$, $R_2R_2R_2R_1$, $R_1R_1R_2R_3$, $R_1R_2R_1R_3$, $R_2R_2R_2R_2$, $R_1R_2R_2R_3$, $R_1R_2R_3R_2$, $R_1R_1R_3R_3$, $R_1R_3R_1R_3$, $R_2R_2R_2R_3$, $R_1R_2R_3R_3$, $R_1R_3R_2R_3$, $R_2R_2R_3R_3$, $R_2R_3R_2R_3$, $R_3R_3R_3R_1$, $R_3R_3R_3R_2$ and $R_3R_3R_3R_3$ can be calculated according to the Formula (6) .

$$\begin{aligned}
P(R_1R_1R_1R_1) &= \frac{C_{N_1}^4}{C_N^4}; \quad P(R_1R_1R_1R_2) = \frac{C_{N_1}^3 \times C_{N_2}^1}{C_N^4}; \\
P(R_1R_1R_2R_2) &= P(R_1R_2R_1R_2) = \frac{1}{2} \times \frac{C_{N_1}^2 \times C_{N_2}^2}{C_N^4}; \\
P(R_1R_1R_1R_3) &= \frac{C_{N_1}^3 \times C_{N_3}^1}{C_N^4}; \quad P(R_2R_2R_2R_1) = \frac{C_{N_2}^3 \times C_{N_1}^1}{C_N^4}; \quad P(R_1R_1R_2R_3) = P(R_1R_2R_1R_3) = \frac{1}{2} \times \frac{C_{N_1}^2 \times C_{N_2}^1 \times C_{N_3}^1}{C_N^4}; \\
P(R_2R_2R_2R_2) &= \frac{C_{N_2}^4}{C_N^4}; \quad P(R_1R_2R_2R_3) = P(R_1R_2R_3R_2) = \frac{1}{2} \times \frac{C_{N_1}^1 \times C_{N_2}^2 \times C_{N_3}^1}{C_N^4}; \quad P(R_1R_1R_3R_3) = P(R_1R_3R_1R_3) = \frac{1}{2} \times \frac{C_{N_1}^2 \times C_{N_3}^2}{C_N^4}; \\
P(R_2R_2R_2R_3) &= \frac{C_{N_2}^3 \times C_{N_3}^1}{C_N^4}; \quad P(R_1R_2R_3R_3) = P(R_1R_3R_2R_3) = \frac{1}{2} \times \frac{C_{N_1}^1 \times C_{N_2}^1 \times C_{N_3}^2}{C_N^4}; \quad P(R_2R_2R_3R_3) = P(R_2R_3R_2R_3) = \frac{1}{2} \times \frac{C_{N_2}^2 \times C_{N_3}^2}{C_N^4}; \\
P(R_3R_3R_3R_1) &= \frac{C_{N_3}^3 \times C_{N_1}^1}{C_N^4}; \quad P(R_3R_3R_3R_2) = \frac{C_{N_3}^3 \times C_{N_2}^1}{C_N^4}; \quad P(R_3R_3R_3R_3) = \frac{C_{N_3}^4}{C_N^4};
\end{aligned}$$

Formula (6)

In formula (6) ,

$$\begin{aligned}
&P(R_1R_1R_1R_1) + P(R_1R_1R_1R_2) + P(R_1R_1R_2R_2) + P(R_1R_2R_1R_2) + P(R_1R_1R_1R_3) + P(R_2R_2R_2R_1) + P(R_1R_1R_2R_3) + \\
&P(R_1R_2R_1R_3) + P(R_2R_2R_2R_2) + P(R_1R_2R_2R_3) + P(R_1R_2R_3R_2) + P(R_1R_1R_3R_3) + P(R_1R_3R_1R_3) + P(R_2R_2R_2R_3) + \\
&P(R_1R_2R_3R_3) + P(R_1R_3R_2R_3) + P(R_2R_2R_3R_3) + P(R_2R_3R_2R_3) + P(R_3R_3R_3R_1) + P(R_3R_3R_3R_2) + P(R_3R_3R_3R_3) = 1
\end{aligned}$$

3.3.2 Probability distribution of four-particle packing gradation

According to the Formula (6) , the probability distribution value P of each packing mode can be calculated. The probability distribution of coal water slurry samples A, B, C and D is shown in Fig.7.

As can be seen from the Fig.7, the probability of four particles forming $R_3R_3R_3R_3$ is the highest in the $N_1N_2N_3$ grading model, which are 57.9%, 56.7%, 56.6%, 55.4% respectively, followed by $R_2R_3R_3R_3$ packing. Under the $N_2N_3N_4$ packing gradation mode, the probability of four particles forming $R_2R_3R_3R_3$ packing is the highest which are 31.5%, 30.6%, 33.6%, 35.5% respectively, followed by $R_3R_3R_3R_3, R_2R_2R_3R_3$ and $R_2R_3R_2R_3$. Under the $N_3N_4N_5$ packing gradation mode, the probability of four particles forming $R_3R_3R_3R_3$ packing is the highest which are 80.2%, 80.0%, 77.8%, 76.6% respectively, followed by $R_2R_3R_3R_3$ and $R_1R_3R_3R_3$.

3.3.3 Distribution of void areas in four-particle packing gradation

According to the calculation value $S_{\max}$ in Formula (5) and the probability value P in formula (6) , the value of $P \times S_{\max}$ in each packing mode can be calculated. The probability distribution of four-particle packing model and the distribution of packing void areas of CWS samples A, B, C and D are shown in Fig. 8.

As can be seen from the Fig.8, the packing void area of four particles forming $R_3R_3R_3R_3$ is the highest in the $N_1N_2N_3$ grading model, which are $6263\mu\text{m}^2, 6134\mu\text{m}^2, 6117\mu\text{m}^2, 5989\mu\text{m}^2$ respectively, followed by $R_2R_3R_3R_3$ packing. Under the $N_2N_3N_4$ packing gradation mode, the packing void area of four particles forming $R_2R_3R_3R_3$ packing is the highest which are $1056\mu\text{m}^2, 1025\mu\text{m}^2, 1127\mu\text{m}^2, 1192\mu\text{m}^2$ respectively, followed by $R_2R_2R_2R_3, R_2R_2R_3R_3$ and $R_2R_3R_2R_3$. And packing modes of $R_3R_3R_3R_3, R_3R_3R_3R_1, R_1R_2R_3R_3, R_1R_3R_2R_3$ also have a larger contribution to packing void area. Under the

$N_3N_4N_5$ packing gradation mode, the probability of four particles forming $R_3R_3R_3R_3$ packing is the highest which are $232\mu m^2, 231\mu m^2, 225\mu m^2, 222\mu m^2$ respectively, followed by $R_2R_3R_3R_3$ and $R_1R_3R_3R_3$. The contribution of the other 18 kinds of packing modes to the packing void area is small, and the accumulative value are $25.8\mu m^2, 26.6\mu m^2, 31.3\mu m^2, 33.1\mu m^2$ respectively.

3.3.4 Cumulative void area of four-particle packing gradation

Based on the calculation of the probability distribution of packing gradation and the area distribution, the cumulative void area of packing gradation $\sum P \times S_{\max}$ is calculated in order to further analyze the difference of packing void in different granular packing states, see Formula (7).

$$\begin{aligned} \sum P \times S_{\max} = & S_{\max}(R_1R_1R_1R_1) \times P(R_1R_1R_1R_1) + S_{\max}(R_1R_1R_1R_2) \times P(R_1R_1R_1R_2) + S_{\max}(R_1R_1R_2R_2) \times P(R_1R_1R_2R_2) + \\ & S_{\max}(R_1R_2R_1R_2) \times P(R_1R_2R_1R_2) + S_{\max}(R_1R_2R_1R_3) \times P(R_1R_2R_1R_3) + S_{\max}(R_2R_2R_2R_1) \times P(R_2R_2R_2R_1) + \\ & S_{\max}(R_1R_1R_2R_3) \times P(R_1R_1R_2R_3) + S_{\max}(R_1R_2R_2R_3) \times P(R_1R_2R_2R_3) + S_{\max}(R_2R_2R_2R_2) \times P(R_2R_2R_2R_2) + \\ & S_{\max}(R_1R_2R_2R_3) \times P(R_1R_2R_2R_3) + S_{\max}(R_1R_2R_3R_2) \times P(R_1R_2R_3R_2) + S_{\max}(R_1R_1R_3R_3) \times P(R_1R_1R_3R_3) + \\ & S_{\max}(R_1R_3R_1R_3) \times P(R_1R_3R_1R_3) + S_{\max}(R_2R_2R_2R_3) \times P(R_2R_2R_2R_3) + S_{\max}(R_1R_2R_3R_3) \times P(R_1R_2R_3R_3) + \\ & S_{\max}(R_1R_3R_2R_3) \times P(R_1R_3R_2R_3) + S_{\max}(R_2R_2R_3R_3) \times P(R_2R_2R_3R_3) + S_{\max}(R_2R_3R_2R_3) \times P(R_2R_3R_2R_3) + \\ & S_{\max}(R_3R_3R_3R_1) \times P(R_3R_3R_3R_1) + S_{\max}(R_3R_3R_3R_2) \times P(R_3R_3R_3R_2) + S_{\max}(R_3R_3R_3R_3) \times P(R_3R_3R_3R_3) \end{aligned} \quad \text{Formula (7)}$$

The cumulative packing void areas of CWS samples A, B, C and D is shown in Fig.9. It can be seen from Fig. 9 that under the condition of $N_1N_2N_3$ packing gradation, the accumulative packing voids area of sample A, B, C and D are $14470\mu m^2, 14606\mu m^2, 14610\mu m^2$ and $14886\mu m^2$ respectively. The accumulated accumulation void area of D sample is the highest. Under the condition of $N_3N_4N_5$ packing gradation, the accumulative packing voids area of sample A, B, C and D are $351\mu m^2, 352\mu m^2, 357\mu m^2, 359\mu m^2$ respectively. The accumulative packing voids area of sample D are the highest. As can be seen from Table 1, the concentration of sample D is the lowest. The results are consistent with the conclusion that the higher the packing efficiency the higher the CWS concentration.

Under the condition of $N_2N_3N_4$ packing gradation, the accumulative packing voids area of sample A, B, C and D are $4761\mu m^2, 4881\mu m^2, 4457\mu m^2, 4094\mu m^2$ respectively. The accumulated void area of D sample is the lowest. The reason is the same as the situation of three-particle packing gradation and it is also that the fine coal powder can not fill the voids formed by medium-size coal powder effectively, which results in the decrease of packing voids and the decrease of slurry concentration.

Therefore, it is feasible to use the larger value of the cumulative probability void area $\sum P \times S_{\max}$ under $N_1N_2N_3$ and $N_3N_4N_5$ packing modes and the smaller value of the cumulative probability void area $\sum P \times S_{\max}$ under $N_2N_3N_4$ packing modes as the evaluation indexes of the CWS concentration reduction. Moreover, the results are basically consistent with the results of comparative analysis of packing voids of three-particle packing gradation. The consistency of the method is further proved.

4. Prediction evaluation procedure for packing efficiency

A process for predicting packing efficiency and slurry concentration based on packing gradation model is as follows:

First step: Calculate the packing void area $S_{\min}$ or $S_{\max}$.

Second step: Calculate the numbers of packing particles per unit volume according to the test value or designed values of particle size distribution.

Third step: Calculate probability of different packing modes by using of Excel softwar according to the formula of permutation and combination.

Fourth step: Calculate the values of $\sum P \times S_{\min}$ or $\sum P \times S_{\max}$ under the packing gradation of $N_1N_2N_3, N_2N_3N_4$ or $N_3N_4N_5$.

Fifth step: Adjust particle size distribution of CWS according to the value of $\sum P \times S_{\min}$ or $\sum P \times S_{\max}$.

Conclusions

(1) The packing efficiency of particles can be quantitatively evaluated by using of cumulative calculation of voids areas and packing probabilities under different packing modes. Correspondently, the CWS concentration can also be predicted by this method.

(2) The slurry concentration of CWS is closely related to its particle distribution. The size of CWS particles determines the size of the voids formed by the packing gradation of particles, and the composition of particles determines the probability of the packing gradation of CWS particles.

Under the condition of wet overflow milling, the average size of slurry calculated by weighted average can reflect the state of packing gradation of slurry particles, which is un-proportionally with the slurry concentration. The larger the average size of slurry, the lower the concentration of CWS slurry, the smaller the average size of slurry, the higher the concentration of CWS slurry.

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Figures

Figure 1

Fig. 1 is the experimental diagram for particles evaluation. The size composition of CWS consists of 8-mesh, 14-mesh, 40-mesh, 80-mesh and 200-mesh. Most of the size composition of qualified CWS from 8 to 14-mesh is less than 1% . Therefore, five particle sizes of 14-40 mesh, 40-80 mesh, 80-200 mesh, 200-325 mesh, <325 Mesh were selected as the research objects. According to the packing model in Fig. 2, the packing voids of particles in CWS can be calculated simply by the distribution of the above five kinds of particle sizes.

Figure 2

Fig.2 is the model of particle packing gradation. In Figure 2, $R_1R_1R_1$, $R_1R_1R_2$, $R_1R_2R_2$, $R_2R_2R_2$, $R_1R_1R_3$, $R_1R_2R_3$, $R_1R_3R_3$, $R_2R_3R_3$ and $R_3R_3R_3$ are packed in a steady state with a radius of R_1 , R_2 , R_3 . $R_1R_1R_1R_1$, $R_1R_1R_1R_2$, $R_1R_1R_2R_2$, $R_1R_2R_1R_2$, $R_1R_1R_1R_3$, $R_1R_2R_2R_1$, $R_1R_1R_2R_3$, $R_1R_2R_1R_3$, $R_2R_2R_2R_2$, $R_1R_2R_2R_3$, $R_1R_2R_3R_2$, $R_1R_1R_3R_3$, $R_1R_3R_1R_3$, $R_2R_2R_2R_3$, $R_1R_2R_3R_3$, $R_1R_3R_2R_3$, $R_2R_2R_3R_3$, $R_2R_3R_2R_3$, $R_3R_3R_3R_1$, $R_3R_3R_3R_2$, $R_3R_3R_3R_3$ are packed in a non-steady state with a radius of R_1 , R_2 , R_3 .

Figure 3

The size of the void formed between particles can be expressed by the area of the triangle formed by the three central points of particles. As shown in Figure 3(a), (b) in the triangle ABC. The area of the triangle can be calculated by Heron's formula

Figure 4

In Fig. 4, N1N2N3 is the packing probability distribution of particles with diameter D1, D2, D3 and number of particles N1, N2, N3. N3N4N5 is the packing probability distribution of particles with diameter D3, D4, D5 and number of particles N3, N4, N5. As can be seen from the diagram, the probability of three particles forming R3R3R3 is the highest in the N1N2N3 grading model, which is 66.4%, 65.3%, 65.2% and 64.2% respectively, followed by R2R3R3 packing. Under the N2N3N4 packing gradation mode, the probability of three particles forming R2R3R3 packing is the highest which are 38.4%, 37.9%, 38.7%, 38.5% respectively, followed by R3R3R3 and R2R2R3. Under the N3N4N5 packing gradation mode, the probability of three particles forming R3R3R3 packing is the highest which is 84.7%, 84.6%, 82.8% and 81.9% respectively, followed by R1R3R3 and R2R3R3. The probabilities of packing modes of R1R1R1, R1R1R2, R1R2R2, R2R2R2, R1R1R3, R1R2R3, R2R2R3 are small and their accumulative probabilities are 0.8%, 0.9%, 1.1% and 1.2% respectively.

Figure 5

As can be seen from the Fig. 5, in the N1N2N3 packing gradation mode, the contribution of R3R3R3 packing of three particles to the void area is the largest, which are 3107μm², 3059μm², 3053μm², 3005μm² respectively, followed by R2R3R3. Under the N2N3N4 packing gradation mode, the contribution of R2R3R3 to the packing void area is the largest, which are 613μm², 606μm², 619μm² and

616 μm^2 respectively, followed by R2R2R3, R1R2R3 and R3R3R3. And the contribution of different CWS samples to the packing void area varies alternately. Under the N3N4N5 packing gradation mode, the contribution of R3R3R3 to the packing void area is the largest, which are 106 μm^2 , 106 μm^2 , 104 μm^2 and 102 μm^2 respectively, followed by R2R3R3 and R1R3R3. The probabilities of packing void area of R1R1R1, R1R1R2, R1R2R2, R2R2R2, R1R1R3, R1R2R3, R2R2R3 are small and their accumulative probabilities are 7.0 μm^2 , 7.2 μm^2 , 8.5 μm^2 and 9.0 μm^2 respectively.

Figure 6

The cumulative packing void areas of CWS samples A, B, C and D is shown in Fig.6.

Figure 7

As can be seen from the Fig.7, the probability of four particles forming R3R3R3R3 is the highest in the $N_1N_2N_3$ grading model, which are 57.9%~56.7%~56.6%~55.4% respectively, followed by R2R3R3R3 packing. Under the $N_2N_3N_4$ packing gradation mode, the probability of four particles forming R2R3R3R3 packing is the highest which are 31.5%~30.6%~33.6%~35.5% respectively~followed by R3R3R3R3, R2R2R3R3 and R2R3R2R3. Under the $N_3N_4N_5$ packing gradation mode, the probability of four particles forming R3R3R3R3 packing is the highest which are 80.2%~80.0%~77.8%~76.6% respectively~followed by R2R3R3R3 and R1R3R3R3.

Figure 8

As can be seen from the Fig.8, the packing void area of four particles forming R3R3R3R3 is the highest in the N1N2N3 grading model, which are $6263\mu m^2$, $6134\mu m^2$, $6117\mu m^2$, $5989\mu m^2$ respectively, followed by R2R3R3R3 packing. Under the N2N3N4 packing gradation mode, the packing void area of four particles forming R2R3R3R3 packing is the highest which are $1056\mu m^2$, $1025\mu m^2$, $1127\mu m^2$, $1192\mu m^2$ respectively followed by R2R2R2R3, R2R2R3R3 and R2R3R2R3. And packing modes of R3R3R3R3, R3R3R3R1, R1R2R3R3, R1R3R2R3 also have a larger contribution to packing void area. Under the N3N4N5 packing gradation mode, the probability of four particles forming R3R3R3R3 packing is the highest which are $232\mu m^2$, $231\mu m^2$, $225\mu m^2$, $222\mu m^2$ respectively followed by R2R3R3R3 and R1R3R3R3. The contribution of the other 18 kinds of packing modes to the packing void area is small, and the accumulative value are $25.8\mu m^2$, $26.6\mu m^2$, $31.3\mu m^2$, $33.1\mu m^2$ respectively

Figure 9

The cumulative packing void areas of CWS samples A, B, C and D is shown in Fig.9.

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