Research Article

Study on Filling Material Ratio and Filling Effect: Taking Coarse Fly Ash and Coal Gangue as the Main Filling Component

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Filling mining is an effective way to settle the dilemma of “Three Down and One Above” in coal mining. Fly ash and coal gangue can be used as filling materials with significant social, economic, and environmental benefits. Using coarse fly ash base as cementing material and coal gangue as aggregate, orthogonal experiment of filling paste was conducted in this study. The range analysis was performed for the strength and transportation requirements of filling paste, and the optimum proportion was determined by the comprehensive balance method. In order to verify the filling effect, a dynamic filling simulation device was designed, and a comparative simulation test of caving mining and dynamic filling mining was carried out. Results show that the filling paste with fly ash and coal gangue as the main component can meet the requirements of filling design and application. This research provides a reference for the material selection and proportion design of paste filling.

1. Introduction

During the underground mining, a large area of voids can be generated when the ore is removed. To maximize ore extraction and the regional ground support, these voids should be backfilled [1, 2]. Filling mining can scientifically reduce the impact and damage of mining on the nature, society, and living environment, maximize the use of limited resources, and realize the “Green Mining” of coal resources [3–5]. Filling mining has become the main technical way to excavate the coal held down under the condition of “Three Down and One Above” in China [6–8]. Paste filling has also been successfully applied in the mining industry of other countries [9–12].

It is of great economic and safety significance to study compressive properties of underground cemented paste backfill (CPB). Stress-strain behaviour of CPB subjected to uniaxial compression and conventional triaxial tests has been discussed widely. The effects of CPB basic components, strength, ageing, and confining pressure on the deformation behaviour of CPB have also been evaluated [13, 14]. Yao and Sun [15] conducted a systematic study to investigate a novel silica alumina-based backfill material composed of coal refuse and fly ash. The results show that the coal refuse and fly ash have different properties under various thermal activation temperatures. Yilmaz et al. [16] analyzed the influence of water cement ratio, cement content, and other factors on the strength of the filling body and studied the influence of consolidation conditions and stress distribution on the physical and mechanical properties of the filling body. Ercikdi et al. [17] studied the influence of material mix ratio on the strength and ultrasonic pulse velocity properties of the filling paste. A FLAC-based numerical model was designed to evaluate the stress state in backfill stopes to ensure the safety of backfilling application [18–20].

With the increasing coal production and stricter regulations on air, water, and ground pollution control, the safety and environmental protections of coal-mine refuse are
demanded urgently. Filling mining may provide an ecological method for the disposal of waste materials [21–24]. For instance, nearly 60% of the electricity in the USA is generated by coal-fired plants, resulting in a large quantity of coal combustion residues which are mainly composed of coal fly ash. Coal fly ash can be reused as typical building materials, such as lime, cement, and gravel, so as to realize the energy conservation and emission reduction [25, 26].

With the change of mining environment, the depletion of superior coal resources and the emergence of government policies on environmental protection, filling mining in coal mines is an indispensable way under certain mining conditions. To reduce the filling cost and increase the comprehensive benefits, the development of filling paste with coarse fly ash and coal gangue as the main component has been deeply studied in this field. However, most research on filling paste is concentrated on the mechanical properties of metal tailings paste. The characteristics of bearing capacity of coal paste and coal mine field transportation have not been considered synthetically. Before the industrial application, there is still a lack of reasonable methods to verify the effect of filling paste. In this paper, coarse fly ash and coal gangue are used as raw materials, orthogonal test is performed to analyze the range of test results, and the optimum proportion of paste filling material is obtained. Through the self-made dynamic filling device, the similar simulation contrast test is conducted between filling and caving mining, and the filling effect is verified finally.

2. Orthogonal Proportioning Test of Filling Paste Material

2.1. Test Materials and Design

2.1.1. Test Material. Daizhuang Coal Mine class III coarse fly ash was used in this test. Microscopic morphology of grade III coarse fly ash particles was observed by a scanning electron microscope (SEM), as shown in Figure 1. These particles are larger, mostly 1~5 μm carbon-bearing spongy angular granules.

In order to improve the early strength of fly ash-based material, a certain proportion of admixture was added, and the mass ratio of fly ash to admixture was 4:1. Admixtures mainly include the following:

(a) Quicklime: the quicklime was purchased from a lime plant with 93% CaO content.

(b) Desulphurized gypsum: desulphurized gypsum is an industrial waste residue removed by wet desulphurization in Huangdao Power Plant. The main component is CaSO4·2H2O.

(c) 32.5 ordinary Portland cement: coal gangues which were taken from Daizhuang coal mine is mainly used as aggregate, and the maximum particle size should be less than 25 mm.

2.1.2. Test Design. According to the strength and transport performance of the paste filling material, the principle of the proportion of the paste filling material is summarized, as shown in Table 1 [27].

The orthogonal analysis method is used in this experiment. The ratio of coarse fly ash-based cementitious material to coal gangue (Factor A), the ratio of coal gangue less than 5 mm (Factor B), and the mass concentration of paste filling material (Factor C) are selected as the three main factors. Factor A ranges from 1:1 to 1:3; Factor B ranges from 30% to 50%; Factor C ranges from 74% to 78%. The three factors were divided into 5 levels. Table 2 shows the orthogonal table L25(5). As shown in Figure 2, the test process includes the following steps: (1) preparing the cuboid test model with side length 100 mm × 100 mm × 100 mm; (2) weighing the corresponding proportion of gangue, cement, fly ash, and water, respectively; (3) mixing the paste filling material evenly and put it into the mixer for full stirring; (4) testing the collapse degree of the mixing uniform paste filling material; (5) testing the mixing uniform paste filling material; (6) putting the mixing material into the standard mold; (7) demoulding after 24 hours; and (8) testing the uniaxial compressive strength of the cured specimen.

2.2. Test Results and Analysis

2.2.1. Range Analysis of Collapse, Stratification, and Bleeding. Table 3 shows the range analysis of collapse, stratification, and bleeding. Table 3 shows the following:

(1) In terms of range R of collapse degree, influence factors on the collapse degree of paste filling materials are ordered as A > C > B, namely, the ratio of coarse fly ash-based cementitious material to coal gangue > the mass concentration of paste filling material > the ratio of coal gangue less than 5 mm. Since the particle size of fly ash is no more than 100 μm basically and the slurry is formed just after being dissolved in water, Factor A has the greatest influence on the collapse degree.

(2) In terms of range R of stratification, influence factors on the stratification of paste filling materials are ordered as C > A > B. The stratification of paste filling material is caused by the relative movement among the different sizes of the paste filling material and uneven distribution of solid particles on the vertical section. If the mass concentration of the paste filling material is small and the water content increases, the downward movement of particles is promoted, resulting in the increase of the stratification degree.

(3) Factors influencing the bleeding rate of paste filling materials are ordered as A > C > B. There are mainly two reasons for water bleeding of paste filling materials. On the one hand, due to the large water content, the excess water cannot be completely absorbed by solid particles, especially the content of cementitious material. On the other hand, the relative movement between the filling materials in the vertical direction leads to the upward movement of water.
Figure 1: Scanning electron micrograph of grade III fly ash. (a) Magnification 1000 times; (b) magnification 2000 times.

Table 1: Proportioning principle of paste filling material.

<table>
<thead>
<tr>
<th>Collapse degree</th>
<th>Stratification degree</th>
<th>8 h strength</th>
<th>28 d strength</th>
<th>Initial setting time</th>
<th>Final setting time</th>
<th>Bleeding rate</th>
<th>Ratio of coal gangue &lt;5 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>&gt;18 cm</td>
<td>&lt;2 cm</td>
<td>&gt;0.1 MPa</td>
<td>&gt;3 MPa</td>
<td>&gt;4 h</td>
<td>&lt;8 h</td>
<td>&lt;3%</td>
<td>35%-45%</td>
</tr>
</tbody>
</table>

Table 2: Orthogonal test with 3 factors and 5 levels L_{25}(5^6).

<table>
<thead>
<tr>
<th>Level</th>
<th>Factor</th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td></td>
<td>1:1</td>
<td>30</td>
<td>74</td>
</tr>
<tr>
<td>2</td>
<td></td>
<td>1:1.5</td>
<td>35</td>
<td>75</td>
</tr>
<tr>
<td>3</td>
<td></td>
<td>1:2</td>
<td>40</td>
<td>76</td>
</tr>
<tr>
<td>4</td>
<td></td>
<td>1:2.5</td>
<td>45</td>
<td>77</td>
</tr>
<tr>
<td>5</td>
<td></td>
<td>1:3</td>
<td>50</td>
<td>78</td>
</tr>
</tbody>
</table>

Figure 2: Test process.
2.2.2. Range Analysis of Compressive Strength. Range analysis of compressive strength is shown in Table 4.

For the sparkline diagram of 8h strength, 1d strength, and 3d strength in Table 4, with the increase of Factor A, the strength tends to decrease in general; with the increase of Factor C, the strength tends to increase. However, there is no obvious correlation between strength and Factor B.

For the sparkline diagram of 7d strength and 28d strength, with the increase of Factor A, the strength decreases generally; with the increase of Factor B, the strength tends to increase. There is no obvious correlation between strength and Factor C.

For the range $R$ of 8h strength, 1d strength, and 3d strength, influence factors on the strength of the paste filling material specimen is ordered as $A > C > B$. Since the hydration of coarse fly ash in water under the action of activator, products such as C-S-H gel and C-A-H, C-A-S-H, and AFt are formed with strong gelation properties. In a certain way, a dense structure is formed by the overlapping and joining of many kinds of gravitation; thus, the strength is generated. In addition, mass concentration also has a certain effect on the early strength. When the mass concentration is small, the early strength is relatively small, and when the mass concentration is high, the early strength is relatively large.

For the range $R$ of 7d strength and 28d strength, the influence factors on the strength of the paste filling material specimen ordered as $A > B > C$ in the later stage. Since the hydration degree of coarse fly ash-based cementitious materials is not complete in the early stage, coarse fly ash-based cementitious materials continue to be hydrated. It can be

### Table 3: Range analysis of paste filling material collapse, stratification, and bleeding.

<table>
<thead>
<tr>
<th>Index</th>
<th>Collapse degree (cm)</th>
<th>Stratification degree (cm)</th>
<th>Bleeding rate (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>B</td>
<td>C</td>
</tr>
<tr>
<td>$k_1$</td>
<td>15.72</td>
<td>21.72</td>
<td>22.94</td>
</tr>
<tr>
<td>$k_2$</td>
<td>20.12</td>
<td>21.40</td>
<td>21.60</td>
</tr>
<tr>
<td>$k_3$</td>
<td>22.48</td>
<td>21.24</td>
<td>21.48</td>
</tr>
<tr>
<td>$k_4$</td>
<td>24.02</td>
<td>20.34</td>
<td>20.52</td>
</tr>
<tr>
<td>$k_5$</td>
<td>22.68</td>
<td>20.32</td>
<td>18.48</td>
</tr>
<tr>
<td>$R$</td>
<td>8.30</td>
<td>1.40</td>
<td>4.46</td>
</tr>
</tbody>
</table>

Sparkline-A

Sparkline-B

Sparkline-C

PSF A C B C A B A C B

PSF = primary and secondary factor.
seen that the content of coarse fly ash-based cementitious materials is the main factor that affects the strength of the filling body in all stages. Factor B has a great influence on the later strength of the filling body. This is mainly caused by the particle gradation of fly ash and broken gangue. It is noted that the average strength also shows a piecewise linear growth pattern, and the demarcation point is 7d, as shown in Figure 3. A better hydration effect of paste filling material can be observed with the fast increase of strength before 7d, indicating that the strength growth in the first 7 days is particularly fast. Therefore, the strength of 8h, 1d, and 3d can be seen as early strength for the paste filling material, and the strength of 7d and 28d as the later stage strength.

2.2.3. Optimum Proportion. The comprehensive balance method is used to compare the results of each factor:

**Factor A.** Firstly, considering the uniaxial compressive strength of the filling body, the 8h strength is greater than 0.1 MPa, 28d strength is required to be greater than 3 MPa, and the proportion of 1 : 3 is excluded as the proportion cannot satisfy the requirement. For collapse degree, the proportion of 1 : 1 is excluded as the proportion cannot meet the requirements. For stratification degree, the proportion of 1 : 2 is excluded as the proportion cannot meet the requirements. In addition, from the economic point of view, the increase of cementing materials definitely leads to the increasing cost, so the proportion of 1 : 2.5 is selected for factor A.

**Factor B.** As the secondary factor shown in the range analysis, the effect of factor B on the collapse degree, stratification degree, bleeding rate, and the early strength of the paste filling material is not obvious. For the later strength, the influence degree is second to Factor A, and with the increase of its proportion, the strength also increases. From the sparkline diagram, there is little difference when Factor B is considered as 45% or 50%. Therefore, Factor B is selected at 45% level for simplicity.

**Factor C.** From the range analysis table, the mass concentration has a great influence on the collapse degree, stratification degree, bleeding rate, and early strength of the paste. When the mass concentration is too small, namely, 74% and 75%, the stratification degree is more than 2 cm, and then the transportation requirement cannot be met; the bleeding rate is also at the edge of the

<p>| Table 4: Range analysis of compressive strength of the paste filling material specimen. |
|---------------------------------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|</p>
<table>
<thead>
<tr>
<th>Index</th>
<th>8h UCS (MPa)</th>
<th>1d UCS (MPa)</th>
<th>3d UCS (MPa)</th>
<th>7d UCS (MPa)</th>
<th>28d UCS (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>k1</td>
<td>0.21</td>
<td>0.19</td>
<td>0.17</td>
<td>0.41</td>
<td>0.40</td>
</tr>
<tr>
<td>k2</td>
<td>0.24</td>
<td>0.19</td>
<td>0.19</td>
<td>0.41</td>
<td>0.41</td>
</tr>
<tr>
<td>k3</td>
<td>0.21</td>
<td>0.21</td>
<td>0.21</td>
<td>0.41</td>
<td>0.40</td>
</tr>
<tr>
<td>k4</td>
<td>0.18</td>
<td>0.21</td>
<td>0.22</td>
<td>0.40</td>
<td>0.40</td>
</tr>
<tr>
<td>k5</td>
<td>0.16</td>
<td>0.20</td>
<td>0.22</td>
<td>0.34</td>
<td>0.41</td>
</tr>
<tr>
<td>S</td>
<td>0.20</td>
<td>0.40</td>
<td>1.19</td>
<td>2.19</td>
<td>3.60</td>
</tr>
<tr>
<td>R</td>
<td>0.08</td>
<td>0.02</td>
<td>0.05</td>
<td>0.09</td>
<td>0.01</td>
</tr>
</tbody>
</table>

\( \bar{S} \) = average uniaxial compressive strength, MPa.
critical value. All levels of initial strength and later strength can meet the requirements; therefore, the optimum concentration is 76% in terms of economic considerations.

According to the research results and previous experience, the optimum proportion of paste filling materials is shown in Table 5.

### 3. Dynamic Simulation Contrast Verification of Paste Filling

#### 3.1. Development of Dynamic Paste Filling Device

How to obtain the simulation effect of dynamic filling is the first problem that must be solved in the similar material test. According to the purpose of the experiment, a dynamic paste filling device was designed to complete the dynamic simulation of the filling process, as shown in Figures 4 and 5.

The dynamic paste filling device is divided into three parts: pressure system, grouting device, and filling groove. The pressure system is made of low carbon steel with a height of 700 mm, a wall thickness of 10 mm, and an inner diameter of 250 mm. The lower part is supported by the base. U-shaped steel is made for two sides of the base. The bottom is a circular iron plate with a wall thickness of 10 mm. The filling groove is made of polyethylene transparent sheets with a thickness of 3 mm and a filling hole diameter of 15 mm. The size of filling groove is 400 mm * 60 mm * 30 mm, which is suitable for the similar simulation test.

Once the filling starts, the top cover is first opened, the paste filling material is poured into the inner cylinder, and the top cover is secured with screws. The loading plate is pressured by the jack in the lower part, and the loading plate is moved upward, so that the paste filling material can flow evenly into the filling groove. There is a pressure gauge on the upper part of the filling pump. According to the reading of the pressure gauge, the strength of the jack pressure is determined, so that filling paste cannot be sprayed from the filling groove due to the excessive pressure during the filling process.

#### 3.2. Test Schemes for Simulation Test

The engineering background is described in reference [8]. The geometric similarity coefficient is 100, and the bulk density similarity coefficient is 1.5. Top weight is applied to compensate for the stress. As shown in Figure 6, to compare the effects of different mining methods, the caving method should be used to mine the coal seam on the left side of the model, and the excavated distance is 70 cm. When the movement of the upper strata is stable, the filling mining method should be used to mine the right side of the model, a 30 cm pillar is set between the left goaf and right filling area. Grid lines are arranged on the surface of the model. The size of the grid lines is 10 cm * 10 cm. The number of grid lines is 10 transverse and 30 longitudinal according to the need of test observation. On the surface of the model, four polyvinyl chloride thin wires are used as the main observation lines from the bottom to the top and ordered as L1, L2, L3, and L4. The stress of the coal pillar is measured in Figure 6.

In Figure 6, there are purple markers 0–7, which are the displacement observation points for caving mining on the left side of the model; green markers P1–P4 of pillar stress observation points; and blue markers 0–20 for filling mining on the right side of the model.

### Table 5: Proportion of paste filling materials.

<table>
<thead>
<tr>
<th>Level</th>
<th>A</th>
<th>B</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>1:2.5</td>
<td>45</td>
<td>76</td>
<td></td>
</tr>
</tbody>
</table>
First, the coal seam on the left side of the model is excavated by the caving method. The excavation step is 6 cm, and the coal seam is excavated three times a day. The displacement of the observation point is observed and recorded.

Filling mining is composed of the following steps:

(a) Excavating coal seams. The length of the open-off-cut hole is 10 cm, the distance of each excavation step is 6 cm, and the filling step distance is 6 cm.

(b) Paste filling material preparation. The paste filling material is prepared in the laboratory according to the tested ratio; the paste filling material is poured into the filling pump.

(c) Filling the goaf. The high-pressure hose is pushed into the filling groove through the filling hole on one side of the filling groove, and the filling is conducted from inside to outside.
3.3. Test Result Analysis

3.3.1. Mining Failure Analysis. Caving mining is shown in Figure 7. It can be seen that caving zone, fracture zone, and bending zone are formed by caving mining. Overburden rock in the caving zone is seriously damaged, and the rock strata in the caving zone are destroyed into smaller bulk-like blocks. Fractures in the fracture zone are mainly divided into vertical through fractures and transverse separation gap. As the mining failure moves upward, the fractures in the fracture zone appear to heal at the bottom and develop in the upper part. Due to the limitation of mining size, the movement of strata is not sufficient, and an obvious separation space is developed finally.

Figure 8 shows the filling process. Figure 9 shows the movement and deformation of overburden strata with the advance of working face. Figure 10 illustrates the final state of caving and filling mining. When filling mining is conducted, under the influence of mining and filling erosion, the immediate roof strata above the coal seam are relatively broken with small breaking distance, and there is almost no transmission of force. When filling mining is 34 m, the main roof above the immediate roof is fractured and continues to be broken in line with certain rules. With the advance of the working face, there is a separation layer between the main roofs. With the advance of mining, the separation layer gradually develops upward. The space of the separated layer decreases gradually. Finally, the separated layer is disappeared.

It can be seen from Figure 10 that the roof of caving mining is seriously damaged, the height of caving zone is large, the fracture zone is obviously developed, and the caving area is compacted. The fracture zone and bending zone are included with small separation space in the filling mining, while the caving zone is not involved.

As shown in Figures 11 and 12, the subsidence of roof mining by the caving method is much larger than that by the filling method. In Figure 12, the subsidence of each survey line is similar because the strata above the main roof are developed in the form of separated strata during the filling mining. When the strata move to a stable state, there is a high compaction degree between the strata, so the subsidence is changed slightly.

Note: Measuring points in Figure 11 are on the caving mining side, and those measuring points in Figure 12 are on the filling mining side.

3.3.2. Pressure Change of Coal Pillar. Figure 13 shows test results of pillar stress after overburden movement is stabilized in the model. The vertical stress value of point P1 on the left side of the pillar is 6.8 MPa, smaller than that of the original rock, indicating that plastic failure of the coal pillar occurs at this point and the integrity of the coal pillar is poor. The vertical stress value of P2 is 31.2 MPa, and the vertical stress value of point P3 is reduced to 21.8 MPa. It can be seen that the load of the coal pillar is reduced by filling mining, and part of the load is shared by the paste filling material. The vertical stress value of P4 is 20 MPa, which indicates that the coal pillar is situated in the elastic stage, and the integrity of the pillar is good. For filling strip mining, the size of the coal pillar can be reduced and the recovery rate of coal can be improved.

3.3.3. Filling Effect Analysis

(1) Interaction between Filling Body and Overburden Rock. Filling body prevents the roof from sinking and inhibits the development of mining fractures. In the filling mining, the fracture of the overlying strata is not obvious, while there are
obvious fractures only in the side of the open-off-cut and the terminal mining line. Since tensile stress on the rock beam exceeds the tensile strength of the rock beam itself, fractures are generated. Compared with the paste filling material of the middle goaf, the compression amount of the coal pillar on the open-off-cut hole side and the terminal mining line is small. So, the deflection of rock strata in the side of the open-off-cut hole and the terminal mining line is relatively large.

(2) Filling Body Provides Lateral Restraint to the Coal Pillar. After filling for a period of time, the deformation on the left side of the coal pillar is greater than that on the right side through the measurement. From the simulation model, the left side of the pillar is relatively soft and part of the coal pillar collapses. The right side of the coal pillar has a good integrity without obvious signs of loosening.

Assumed that the side pressure of the coal pillar in filling mining is $\sigma_1$, the width of the coal pillar is unchanged, and the increasing bearing capacity of the pillar is $\Delta\sigma$:
\[ \Delta \sigma = (\sigma_c + K \sigma_f ) \left( \frac{0.64 + 0.36W}{m} \right)^{1.4} - \sigma_c \left( \frac{0.64 + 0.36W}{m} \right)^{1.4} = K \sigma_f \left( \frac{0.64 + 0.36W}{m} \right)^{1.4} \]  

(1)

where \( \sigma_c \) is the uniaxial compressive strength when side pressure equals to 0, MPa; \( K \) is the coefficient of confining pressure effect; \( W \) is the width of the coal pillar, m; and \( m \) is the height of the coal pillar, m. Obviously, filling mining can greatly improve the stability of the coal pillar [28, 29].

In summary, the paste filling material changed the supporting system of the goaf, as shown in Figure 14, and the original supporting system of the coalface-coal pillar was changed into that of the coalface-paste filling material-coal pillar. The stress state of the coal pillar has been changed. The unidirectional force is changed into the three-direction force, which greatly improves the bearing capacity of the coal pillar and ensures the stability of the overlying strata.

4. Conclusion

(1) The content of coarse fly ash base cementing material is the main factor that affects the strength of the filling body in the early and later stages. The ratio of coal gangue less than 5 mm has a great influence on the later strength of the filling body. The ratio of 1:2.5 (coarse fly ash-based cementitious material to coal gangue), the mass concentration of 76\%, and the ratio of 45\% (coal gangue less than 5 mm) can meet the requirements of engineering transportation and strength and can be used as the optimum parameters of this experiment.

(2) A dynamic filling device was designed to simulate the filling process. A comparative test is carried out between caving mining and filling mining. Only fracture zones and bending subsidence zones are included in backfill mining. In the filling mining, the immediate roof is transformed into the main roof, and the strata above the main roof top are developed upward in the form of separated layer. The paste filling material supports the overburden, prevents it from sinking, collapses, and provides lateral restraint to the coal pillar.

(3) It is necessary to keep a certain width of the coal pillar at the open-off-cut hole for filling and recovering the isolated island coal pillar. In the early stage of filling mining, the compressive strength of the paste filling material is relatively small. A certain width of the coal pillar plays an important supporting role for the roof; namely, the valuable time can be provided for the consolidation of the strength of the paste filling material. The remaining pillar shares part of the pressure and prevents the paste filling material from flowing out.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare no conflicts of interest.
Authors’ Contributions

All the authors contributed to publishing this paper. C. W. and Yin. L. conceived the main idea of the paper; Yao. L. analyzed the data; C. W. wrote the paper; Y. Y. L. and H. H. did a lot of work to modify figures and proofread the revised version.

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