Process Optimization Experiment of Preparing Light Ceramsite from Qinghai Reshui Coal Gangue

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Abstract. Qinghai has abundant coal resources, but the accumulation of coal gangue poses a certain threat to the fragile ecology of the area. In this paper, coal gangue light ceramsite was prepared by using coal gangue from Qinghai Reshui coal mine as raw material and sodium carbonate as auxiliary material. The optimized process conditions are as follows: the mass ratio coal of gangue and sodium carbonate = 50:1 (10% hydrochloric acid contamination); the drying temperature is 105±2°C; the drying time is 90min; preheating temperature is 450°C; the preheating time is 10 min; the calcination temperature is 950 °C; the roasting time is 15 min. The ceramsite prepared under this condition has a packing density of 423 kg/m³ and a bulk density of 889 kg/m³. The process does not use fly ash, and all the coal gangue raw materials were used without any residue, which can effectively reduce the environmental pressure brought by the accumulation of coal gangue, and has high industrial application value.

1. Introduction

Coal gangue is a kind of solid waste mainly composed of silicon and aluminum produced in the process of coal mining and washing. It is one of the industrial wastes with the largest annual average emissions and accumulated stock in China [1]. The landfill or open-air stacking treatment of coal gangue encroached on a large amount of land, and the environment was seriously polluted due to the effects of drenching and dusting, which is called as "misplaced resources" [2]. The coal in Reshui and Muli of Qinghai province is natural coking coal of low-phosphorus, low-ash and high-quality, and the coal gangue produced by washing has the characteristics of low phosphorus and no heavy metal. Due to the fragile ecological environment of Qinghai Plateau pastoral area, the recycling of solid waste in the Qaidam circular economy industrial circle has increased to new requirements. The ceramsite produced by coal gangue can be applied to the light aggregate of buildings or the adsorption materials of waste liquid treatment [3]. In this work, the coal gangue was crushed, ground, added with auxiliary materials, and finally burned into ceramsite. The effect of foaming process on the ceramsite packing density was investigated.
2. Experimental

2.1. Experimental Materials

The coal gangue used in the experiment is from coal gangue in Haibeizhou Reshui Coal Industry Park in Qinghai Province. It is a clay rock vermiculite such as carbon shale and carbonaceous mudstone, which is suitable for the production of building materials and adsorption materials. The sampling method is the plum sampling method. Table 1 shows the chemical composition of the coal gangue by XRD diffraction analysis.

| Ingredient               | MgO  | Al₂O₃ | SiO₂   | Fe₂O₃ | K₂O   | SO₃   | CaO  | TiO₂ |
|--------------------------|------|-------|--------|-------|-------|-------|------|------|
|                          | (%)  |       |        |       |       |       |      |      |
| MgO                      | 0.106|       |        |       |       |       |      |      |
| Al₂O₃                    | 23.912|      |        |       |       |       |      |      |
| SiO₂                     | 61.122|      |        |       |       |       |      |      |
| Fe₂O₃                    | 6.211 |      |        |       |       |       |      |      |
| K₂O                      | 2.868 |      |        |       |       |       |      |      |
| SO₃                      | 2.705 |      |        |       |       |       |      |      |
| CaO                      | 1.587 |      |        |       |       |       |      |      |
| TiO₂                     | 1.131 |      |        |       |       |       |      |      |

2.2. Experimental Methods

There are three processes in the production process of coal gangue ceramsite, namely raw material processing, granulation and hot processing [4]. First, the coal gangue was crushed, ground and sifted to less than 100 mesh. After stirring with appropriate foaming agent and water, the coal gangue was made into spherical raw material balls between 10mm and 18mm by hand. The spherical raw material balls were dried at 105°C for 90min and then placed in the muffle furnace at 350°C preheated for 10min. Then they were roasted at 1000°C for 10min and cooled to 280°C. Then they were taken out and cooled to room temperature to obtain the finished ceramsite.

3. Results and discussion

3.1. Selection of Foaming Agent and Ratio

The chemical composition of coal gangue is similar to that of clay minerals, which can meet the basic requirements for the preparation of ceramsite. In this paper, coal gangue was used as the main raw material, and different foaming agents are used instead of fly ash as auxiliary materials to prepare ceramsite. The effects of different foaming processes on the coal gangue ceramsite bulk density are investigated. The results of foaming agent selection and proportioning test are shown in Table 2.

| NO | Types                | Content (Wt%) | Processing Method         | ρ(kg/m³) |
|----|----------------------|--------------|--------------------------|---------|
| 1  | Na₂CO₃               | 0.5          | /                        | 573.6   |
| 2  | Na₂CO₃               | 1.0          | /                        | 572.7   |
| 3  | Na₂CO₃               | 2.0          | /                        | 541.5   |
| 4  | Na₂CO₃               | 0.5          | HCl contamination        | 568.1   |
| 5  | Na₂CO₃               | 1.0          | HCl contamination        | 543.6   |
| 6  | Na₂CO₃               | 2.0          | HCl contamination        | 479.3   |
| 7  | NaHCO₃               | 0.5          | /                        | 666.5   |
| 8  | NaHCO₃               | 1.0          | /                        | 625.5   |
| 9  | NaHCO₃               | 2.0          | /                        | 611.9   |
| 10 | NaHCO₃               | 2.0          | HCl contamination        | 514     |
| 11 | CaO                  | 1.0          | Unformed                 | /       |
| 12 | CaO                  | 5.0          | Fragile                  | /       |
| 13 | CaO                  | 9.0          | Fragile                  | /       |
| 14 | Aluminum powder paste| 0.1          | 0.375 Wt% NaOH           | 695.8   |
| 15 | Aluminum powder paste| 0.5          | 1.5 Wt% NaOH             | 667.5   |
| 16 | Aluminum powder paste| 1.0          | 3 Wt% NaOH               | 654.8   |
| 17 | /                    | /            | Blank sample             | 730.9   |
| 18 | /                    | /            | Blank sample             | 737.5   |
| 19 | /                    | /            | Blank sample             | 757.0   |

Table 2. The selection of foaming agent and proportioning test

It can be seen from table 2 that the packing density of ceramsite prepared by using pure coal gangue (no foaming agent) is between 730-760 kg/m³, and its volumetric density is all over 1500kg/m³. The packing density of ceramsite prepared by adding foaming agent into the raw material is obviously
reduced. Under the same test conditions, the foaming effect of sodium carbonate and hydrochloric acid contamination is the best, with a packing density of less than 500 kg/m$^3$ cubed. Therefore, sodium carbonate and hydrochloric acid contamination is selected as the foaming agent. It can also be seen from table 2 that as the content of foaming agent increases, the packing density and volume density of products also decrease correspondingly. At the addition amount of foaming agent is 0.5% and 1.0%, the bulk density does not decrease significantly, while when the addition amount of foaming agent is 2.0%, the packing density changes significantly, indicating that the coal gangue with the addition of 2.0% foaming agent can achieve better results. Considering the economic cost, the adding amount of foaming agent is set at 2.0%, which is far less than the commonly used 5% [5-7].

3.2. Results of Orthogonal Experiments

Orthogonal experimental design is a design method to study multi-factor and multi-level. It is based on the orthogonality to select some representative points from the comprehensive experiment to test, not only can effectively save the experiment time, but also can infer a rigorous and representative best solution. Under the premise of determining the type and amount of foaming agent, the heat treatment process of ceramsite was optimized, and the effects of preheating temperature, preheating time, calcination temperature and calcination time on the properties of ceramsite were investigated. An orthogonal test chart L$_{16}$ (4$^4$) as shown in Table 3 was designed.

| Factors | Preheating Temperature (°C) | Preheating Time (min) | Calcination Temperature (°C) | Calcination Time (min) |
|---------|-----------------------------|-----------------------|-------------------------------|------------------------|
| Levels  | A                           | B                     | C                             | D                      |
| 1       | 350                         | 10                    | 900                           | 5                      |
| 2       | 450                         | 15                    | 950                           | 10                     |
| 3       | 500                         | 20                    | 1000                          | 15                     |
| 4       | 550                         | 25                    | 1050                          | 20                     |

The preheating and roasting of ceramic samples are carried out in the roasting furnace. The influences of preheating temperature, preheating time, roasting temperature and roasting time on the performance parameters of the samples can be determined according to the following orthogonal test of roasting. The orthogonal test results are shown in Table 4.

| NO. | Preheating Temperature (°C) | Preheating Time (min) | Calcination Temperature (°C) | Calcination Time (min) | ρ(kg/m$^3$) |
|-----|-----------------------------|-----------------------|-------------------------------|------------------------|-------------|
| 1   | A                           | 1                     | 1                             | 1                      | 509.4       |
| 2   | 2                           | 2                     | 2                             | 2                      | 495.1       |
| 3   | 3                           | 3                     | 3                             | 3                      | 477.3       |
| 4   | 4                           | 4                     | 4                             | 4                      | 533.2       |
| 5   | 2                           | 1                     | 2                             | 3                      | 444.0       |
| 6   | 2                           | 2                     | 1                             | 4                      | 520.6       |
| 7   | 2                           | 2                     | 4                             | 1                      | 534.8       |
| 8   | 2                           | 4                     | 3                             | 2                      | 532.4       |
| 9   | 3                           | 3                     | 1                             | 4                      | 512.6       |
| 10  | 3                           | 2                     | 4                             | 3                      | 585.3       |
| 11  | 3                           | 3                     | 1                             | 2                      | 510.8       |
| 12  | 3                           | 4                     | 2                             | 1                      | 473.4       |
| 13  | 4                           | 1                     | 4                             | 2                      | 528.4       |
| 14  | 4                           | 2                     | 3                             | 1                      | 538.7       |
| 15  | 4                           | 3                     | 2                             | 4                      | 531.3       |
| 16  | 4                           | 4                     | 1                             | 3                      | 544.7       |

K$_1$: 503.75  498.6  521.375  514.075
K$_2$: 507.95  534.925  485.95  516.675
K$_3$: 520.525  513.55  515.25  512.825
K$_4$: 535.775  520.925  545.425  524.425
R: 32.025  36.325  59.475  11.600
In the orthogonal test table, $K_i$ is defined as the sum of the evaluation indicators at different levels (1, 2, 3, 4) in each of the influencing factors (A, B, C, D). When $K_i$ is minimum, the optimal level of each factor can be obtained [16]. $R$ stands for the extreme difference and is used to assess the extent to which different factors influence the outcome, i.e. a larger $R$ value means a more important factor. According to the experimental results intuitively analyzed in table 4, the packing density of ceramsite prepared by 16 groups of experiments is less than 600 kg/m$^3$. According to $K_i$ values at various levels, we obtained that the optimal process parameter was $A_2B_1C_2D_3$, and the packing density of the prepared ceramsite samples was 444 kg/m$^3$, and the volume density was 1031 kg/m$^3$, which belonged to lightweight aggregate ceramsite. By comparing the $R$ value of each influencing factor, it can be found that the significance order of the influence of each factor on the bulk density of ceramsite is $C>B>A>D$.

| Factors | Type III Sum of squares | df | F  | F-threshold |
|---------|-------------------------|----|----|-------------|
| A       | 2489.087                | 3  | 0.781 | 3.490       |
| B       | 2752.192                | 3  | 0.864 | 3.490       |
| C       | 7172.817                | 3  | 2.253 | 3.490       |
| D       | 325.262                 | 3  | 0.102 | 3.490       |
| Error   | 12736.36                | 12 |     |             |

From the results of the variance analysis in Table 5, it can be seen that $F_c = 2.253 > F_b = 0.864 > Fa = 0.781 > Fd = 0.102$, indicating that the C factor (calcination temperature) has a significant influence on the packing density. Combining the above analysis results and considering the energy saving factors, the optimum conditions could be expressed as $A_2B_1C_2D_3$, that is, the preheating temperature was 450 °C, the preheating time was 10 min, the calcination temperature was 950 °C, and the roasting time was 15 min. Under this process conditions, ceramsite with a packing density of 423 kg/m$^3$ and a bulk density of 889 kg/m$^3$ was prepared, and the calcination temperature was also lower than that of the conventional process [5-7].

4. Conclusion
Coal gangue in Qinghai Reshui can be used as industrial raw material to produce coal gangue ceramsite. The addition of foaming agent can effectively reduce the packing density of ceramsite. The method of taking sodium carbonate as foaming agent and 10% hydrochloric acid solution as contamination has the best effect, and its process raw material ratio is coal gangue: sodium carbonate =100:2.

Through orthogonal experiment optimization, the effect of roasting temperature on the bulk density was the greatest. The optimum process conditions for the production of coal gangue ceramsite are: preheating temperature 450 °C, preheating time 10 min, calcination temperature 950 °C, and the roasting time 15 min. The ceramsite bulk density 423 kg/m$^3$. According to the experimental results, the process has the advantages of low amount of foaming agent, low roasting temperature and no need to add fly ash.

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