Formation characteristics of ultra fine particles during oxy-coal combustion with flue gas recirculation

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Abstract: A laboratory-scale oxy-fuel combustion experimental apparatus was built up to simulate the real oxy-fuel combustion. Under conditions with three kinds of recycles (no-RFG, de-ash and ash), combustion experiments of DT bituminous coal (rich in Si and Al) and SH bituminous coal (rich in Ca) were conducted to investigate the influence of recycled ash on formation of ultrafine particle matters. The results show that, compared with the no-RFG condition, under condition with de-ash flue gas recycle, the yields of ultrafine particle matters of the DT and SH coal were both basically invariant, while that under condition with ash recycle decreased by about 12.03% (DT coal) and 7.73% (SH coal), respectively. Under condition with ash recycle, the silicon aluminate, which was rich in the ash of DT coal, would absorb the Na and K vapor released during the combustion, and the rich CaO in the SH coal may also absorb the S element during the combustion to restrain the S from migrating to the ultrafine particles, both of the reactions decreased the formation of the ultrafine particles.

1. Introduction

Oxygen-enriched (O₂/CO₂) combustion is a new type of combustion technology in which the fuel is burned in an atmosphere consisting of pure oxygen and recycled flue gas. Oxygen-enriched combustion can be achieved by adjusting the ratio of O₂ to circulating flue gas. Achieve the same heat and mass transfer effect as traditional air combustion². According to the degree of purification of circulating flue gas, the flue gas cycle can be divided into clean flue gas circulation and a dust-containing flue gas circulation. Compared with conventional air combustion, the combustion characteristics of pulverized coal, the composition of flue gas, the conversion of minerals, the accumulation of ash and slag, and the generation of pollutants have changed. At present, the research on the formation characteristics of particulate matter in the oxyfuel combustion mode of coal powder is mostly a single-channel experiment, that is, the O₂/CO₂ gas distribution is used to simulate the oxyfuel combustion, and it is not the true sense of flue gas circulation oxyfuel combustion.

In this paper, a flue gas circulation combustion system simulating real oxy-combustion combustion was built on the basis of laboratory high temperature settling furnace (DTF) system, and the amount of granules generated by ash-containing flue gas circulation and de-ash flue gas circulation was studied. At the same time, by comparing the difference of particle formation between two different minerals, the influence of the main components (Si, Al and Ca) in the circulating ash on the formation characteristics of ultrafine particles during the cycle is discussed.
2. Experimental Part

2.1. Coal sample and its low temperature ash analysis
The experiment uses Datong (DT) bituminous coal and Shenhua (SH) bituminous coal. The coal sample industry and elemental analysis results are shown in Table 1. The results of X-ray fluorescence (XRF) analysis of low temperature ash are shown in Table 2. It can be seen that the low-temperature ash Si content of DT coal is higher, while the low-temperature ash Ca content of SH coal is higher.

| Coal Sample | Industrial analysis | Elemental analysis |
|-------------|---------------------|-------------------|
|             | Mar | Aar | Var | FCar | Cdaf | Hdaf | Ndaf | Sdaf | O*daf |     |
| DT Coal     | 2.74| 37.74| 27.22| 32.3 | 83.78| 6.01 | 1.18 | 0.36 | 8.67 |     |
| SH Coal     | 1.84| 21.41| 24.89| 51.86| 85.35| 3.71 | 1.35 | 0.56 | 9.03 |     |

| Table 2. Component analysis for low temperature ash of DT coal and SH coal     w% |
|------------------------------------------|
| Coal Sample | Al₂O₃ | SiO₂ | CaO | Fe₂O₃ | MgO | Na₂O | SO₃ | K₂O | P₂O₅ | ZnO | MnO |
| DT Coal     | 33.64 | 45.09 | 1.98 | 3.88 | 1.77 | 2.94 | 7.67 | 0.45 | 0.72 | 0.02 | 0.03 |
| SH Coal     | 34.52 | 37.40 | 7.08 | 4.61 | 1.85 | 2.79 | 8.05 | 1.22 | 1.29 | 0.02 | 0.06 |

2.2. Experimental system
The experimental system is built on the basis of a settling furnace (DTF). The new flue gas circulation system includes: flue gas circulation pump, stainless steel pipeline, pipeline heating system, vacuum gauge, valve, flowmeter, and flue gas analyzer. The circulation line is kept at 200 °C to prevent condensation of water vapor and SO₂, SO₃ and other gases in the flue gas.

2.3. Experimental conditions
The experimental atmosphere includes no-flame gas circulation (no-RFG, ie O₂/CO₂), de-ash and ash-containing flue gas circulation (ash). The experimental conditions are shown in Table 3. In Table 3: Q₀ is the gas flow rate in the furnace, Q₁ is the distribution gas flow directly from the cylinder, and Q₂ is the circulating flue gas flow. The gas composition basically maintains O₂/CO₂=30/70 under all working conditions. Define the cycle ratio as the ratio of the circulating flue gas flow to the furnace gas flow, ie ϕ=Q₂/Q₀. The cycle ratio in all flue gas cycle conditions is set to ϕ=0.6, which is similar to the existing large flue gas recirculation test device.

| Serial number | Coal | Loop type | Gas flow (/L·min⁻¹) | Cycle ratio |
|---------------|------|-----------|---------------------|-------------|
|               |      |           | Q₀      | Q₁ | Q₂ |      |
| 1             | DT   | no-RFG    | 5       | 5 | 0  | 0    |
| 2             | DT   | de-ash    | 5       | 2 | 3  | 0.6  |
| 3             | DT   | ash       | 5       | 2 | 3  | 0.6  |
| 4             | SH   | no-RFG    | 5       | 5 | 0  | 0    |
| 5             | SH   | de-ash    | 5       | 2 | 3  | 0.6  |
| 6             | SH   | ash       | 5       | 2 | 3  | 0.6  |

The experimental temperature was set to 1500 °C in all working conditions, the gas flow rate in the furnace was 5 L/min, and the residence time of the combustion products in the furnace was about 1.2 s. During the experiment, the gas is simulated by the primary air, and the pulverized coal is sent to the furnace for combustion. The powder feeding rate is 0.2 g/min. In the deashing flue gas cycle experiment, a filtering device is added to the circulating branch to remove particulate matter in the flue gas, and only the gaseous flue gas component is allowed to be recycled back to the furnace.
3. Results and discussion

3.1. Mass particle size distribution of PM
The mass particle size distribution of the two coals burning under different flue gas conditions is shown in Fig 1. It can be seen from Fig 1, that PM$_{10}$ produced by combustion of two coals in O$_2$/CO$_2$ atmosphere has a bimodal particle size distribution, including ultrafine mode with peak particle size of ~0.08 $\mu$m and peak particle size of ~5 $\mu$m. When the combustion atmosphere changes from O$_2$/CO$_2$ to deashing flue gas cycle, PM$_{10}$ produced by DT and SH coal combustion still has a bimodal distribution, indicating that the formation mechanism of PM$_{10}$ in O$_2$/CO$_2$ atmosphere and deashing flue gas circulation atmosphere is not significant difference, which is consistent with the findings of Sheng et al[3]. In the de-ash flue gas cycle, most of the flue gas entering the furnace is CO$_2$ and residual O$_2$ produced by burning coal. The gas composition of the combustion environment is constant and only the relative content of each component changes, so the formation mechanism of the particulate matter is not change.

![Fig 1. Particle size distribution of PM$_{10}$ formed during combustion of DT and SH coal](image)

3.2. Influence of different atmospheres on the formation of ultrafine particles
The PM$_{0.2}$ production amount of DT coal and SH coal under various experimental conditions is shown in Fig 2. It can be seen from Fig 2, that under the O$_2$/CO$_2$ atmosphere and the de-ash flue gas circulation condition, after the DT coal is burned, the mass of PM$_{0.2}$ generated per 1 g of ash is 32.33 and 31.34 mg, and there is no significant difference between them, SH coal is similar. Under the above two combustion conditions, there is no additional fly ash particles in the flue gas, and the formation of ultrafine particles is mainly determined by the gasification behavior of the minerals in the coal. The gasification behavior of minerals is controlled by the combustion temperature and O$_2$ content in the combustion atmosphere[4-7]. Since the furnace wall temperature is a uniform set temperature in the experiment, the combustion temperature of the coal char is mainly determined by the O$_2$ content in the atmosphere. Figure 3 shows the O$_2$ volume fraction of the furnace inlet gas under various working conditions. It can be seen that the O$_2$ volume fraction of the flue gas entering the furnace under each working condition has no significant change, so the amount of ultrafine particulate matter in the deashing flue gas circulation has no significant change.
Fig 2. The yield of PM$_{0.2}$ formed during combustion of DT coal and SH coal

However, under the ash-containing flue gas combustion conditions, the PM$_{0.2}$ production of DT coal and SH coal decreased by about 12.03% and 7.73%, respectively, which may be caused by fly ash carried in the circulating flue gas.

3.3. Effect of circulating ash properties on the formation of ultrafine particles

Figure 4 shows the XRD results of total fly ash samples after DT and SH coal combustion. It can be seen that the ash cycle basically does not change the mineral composition of coal-fired fly ash, which is consistent with the experimental results of Wang et al.[8,9]. It is worth noting that there is a large amount of mullite and SiO$_2$ in the total fly ash produced by the combustion of DT coal. Since the circulating ash has a long residence time in the furnace, the minerals such as aluminosilicate and SiO$_2$ may capture and fix alkali metal vapors such as Na and K produced by burning coal, thereby reducing the amount of PM$_{0.2}$ generated.

4. Conclusions

(1) Under the three combustion conditions of smokeless gas cycle (O$_2$/CO$_2$), de-ash flue gas cycle and ash-containing flue gas cycle, the mechanism of pulverized coal combustion to produce ultrafine particles is the same. Under the first two conditions, there is no significant change in the amount of ultrafine particles produced by the combustion of DT coal and SH coal.

(2) For DT coal with high silicon and aluminum content, after adding ash cycle, the aluminosilicate in the ash will absorb the alkali metal vapors such as Na and K released during the coal combustion.
process, and inhibit the gasification-condensation method to the ultrafine particles. Thereby inhibiting the production of PM$_{0.2}$.

(3) For SH coal with high Ca content, after adding ash cycle, in addition of aluminosilicate in ash inhibiting PM$_{0.2}$ formation, the CaO content in the circulating ash will also absorb the S element released from coal combustion, reducing its conversion to ultrafine particle, thereby further reducing the amount of PM$_{0.2}$ generated.

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