Temperature Effect on Central-Mode Particulate Matter Formation in Combustion of Coals with Different Mineral Compositions

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ABSTRACT: Coal combustion and mineral particle heating experiments were carried out in a drop-tube furnace at 1373 and 1573 K, respectively, to investigate the temperature effect on central-mode particulate matter (PM) formation during the combustion of coals with different mineral compositions. Two bituminous coals, coal A and coal B, with similar organic properties but different Ca/Fe mineral contents were tested. Typical minerals in the two coals, calcite and kaolinite, were used in the mineral particle heating experiments. An air atmosphere, a sample-feeding rate of 0.3 g/min, and a particle residence time of about 2 s were adopted in these experiments. The PM and bulk ash samples were collected by a low-pressure impactor and fiber filters, respectively, through a water-cooled N2-quenched probe. The elemental compositions, mass concentrations of PMs, mineral compositions, and morphologies of bulk ashes were characterized. The results show that the mass fraction size distribution of aluminum (Al) can be used to identify the different PM formation modes. When the temperature is increased from 1373 to 1573 K, the central-mode PM concentration for coal A increases by 61.8%, whereas that for coal B decreases by 13.2%. The remarkable difference is attributed to different fragmentation and coalescence behaviors resulting from different mineral compositions of the two coals. The criteria of optimal coal mineral composition for melting-phase generation and coalescence occurrence are developed. Interactions between calcite and kaolinite and their influence on central-mode particle formation with respect to the temperature are clarified by the mineral particle heating experiments.

1. INTRODUCTION

Particulate matter with an aerodynamic diameter equal to or smaller than 2.5 μm (PM$_{2.5}$) is a major air pollutant in most Chinese cities, and coal-fired power plants are an important PM$_{2.5}$ source. Figure 1 shows the formation mechanisms of coal-derived particulate matter (PM). Dependent upon its formation mechanisms, PM can be divided into three formation modes: ultrafine mode, coarse mode, and a newly reported fine mode.5 The ultrafine-mode particles are mainly formed by the vaporization–condensation mechanism;6,7 the coarse-mode particles are mainly formed by char fragmentation and mineral coalescence;7,8 and the central-mode particles are likely formed by fragmentation mechanisms, including char fragmentation,9,10 mineral fragmentation,9,11,12 etc. and direct transformation of the fine particles originally contained in raw coal.9,13,14 The ultrafine-mode PM and part of the central-mode PM constitute the coal-derived PM$_{2.5}$. However, in comparison to the well-known ultrafine mode, the formation of PM in central mode is far less understood and warrants further investigation.

The reaction temperature has a significant influence on PM formation during coal combustion. Numerous investigations have shown that the PM concentration tends to increase with an increase in the temperature.15,16 However, available research on the effect of the temperature on central-mode PM formation led to different conclusions. By burning a size-classified Chinese bituminous coal, Yu et al.3 observed that the central-mode PM concentration increased significantly when the temperature was increased from 1373 to 1673 K. However, during the combustion of an American bituminous coal, Fix et al.17 found that the central-mode PM formation decreased slightly when the peak temperature was increased from 1508 to 1606 K. Buhre et al.18 investigated five Australian coals and found that the PM$_{2.5}$ (central-mode PM was an important part) concentration displayed different variation tendencies with an increase in the temperature; it showed an obvious increase for four coals, whereas it remained constant for another coal (considering the increase in the ultrafine-mode PM concentration, the central-mode PM concentration for this coal decreased to some extent). The causes for the different results were not explored in the aforementioned literature. Never-
theless, a careful examination of the data reported indicates that
differences in the content of Ca/Fe species in the coals may
result in the phenomenon that the central-mode PM
centre experiences opposite variation tendencies with
an increase in the temperature. Wang et al.19 found that Ca/Fe
species could react with aluminosilicates in coal and influence
the formation of particulate matter with an aerodynamic
diameter equal to or larger than 1 μm (PM1+) generated during
combustion. However, the relationship between the temper-
ature effect on central-mode PM formation and coal mineral
composition (especially the Ca/Fe mineral contents) has not
been considered in the literature.

This work aims to understand the temperature effect on
central-mode PM formation by varying coal mineral
composition. First, two bituminous coals with similar organic
properties but different Ca/Fe mineral contents were burned in
a drop-tube furnace (DTF) at 1373 and 1573 K, respectively.
The variation tendencies of the central-mode PM concen-
trations with an increase in the temperature were compared,
and the influence of the mineral composition was discussed.
Second, mineral particle heating experiments using typical
minerals present in the coals were carried out to clarify the
mineral interactions and their effect on central-mode particle
formation under the conditions of coal combustion.

2. EXPERIMENTAL SECTION

2.1. Sample Preparation and Analysis. Two Chinese
bituminous coals, coal A and coal B, with their basic properties
shown in Table 1, were used in the coal combustion experiments. The
organic properties of the two coals, including volatile matter (31.87
versus 28.87%), fixed carbon (56.22 versus 54.03%), ultimate
elemental composition and vitrinite content (50.5 versus 53.4%), are
quite similar. However, the inorganic compositions detected by X-ray
fluorescence (XRF), including Al2O3 (14.46 versus 24.82%), SiO2
(31.96 versus 41.52%), CaO (22.03 versus 11.42%), and Fe2O3 (10.77
versus 6.48%), vary significantly between the two coals. The
characteristics of the minerals in the two coals were quantified using
computer-controlled scanning electron microscopy (CCSEM),20 and
the results are shown in Figures 2−4. Figure 2 shows the mineral
compositions of coals A and B: (a) coal A and (b) coal B.

Figure 2. Mineral compositions of coals A and B: (a) coal A and (b) coal B.

The proportions of excluded minerals in the two coals are very similar (76.8% in coal A and 76.0% in coal B).
Calcite and kaolinite were selected, blended, and heated in mineral
particle heating experiments to elucidate the effect of mineral
interactions on central-mode particle formation. Calcite was selected
rather than iron oxide/siderite for two reasons: first, the effect of Ca
and Fe minerals on PM formation are thought to be similar,19 and
second, because of the limitations of CCSEM analysis, iron oxide and
siderite cannot be effectively identified. The purity of the calcite used
was >98%. The composition of kaolinite used is shown in Table 1. The
ratio CaO/(Al2O3 + SiO2) for the blended mineral (calcite + kaolinite)
sample was the same as that for coal B. All of the samples, coals and
minerals, were ground and sieved to 45−100 μm in particle size.

2.2. Experimental Conditions, Ash Sampling, and Analysis. Coal combustion and mineral particle heating experiments were

Table 1. Properties of the Samples Used

<table>
<thead>
<tr>
<th>proximate analysis (wt %, air-dried basis)</th>
<th>major ash components (wt %)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>coal A</td>
</tr>
<tr>
<td>moisture</td>
<td>4.64</td>
</tr>
<tr>
<td>ash</td>
<td>7.27</td>
</tr>
<tr>
<td>volatile matter</td>
<td>31.87</td>
</tr>
<tr>
<td>fixed carbon</td>
<td>56.22</td>
</tr>
<tr>
<td>ultimate analysis (wt %, air-dried basis)</td>
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</tr>
<tr>
<td>carbon</td>
<td>45.72</td>
</tr>
<tr>
<td>hydrogen</td>
<td>3.84</td>
</tr>
<tr>
<td>nitrogen</td>
<td>0.81</td>
</tr>
<tr>
<td>vitrinite content (wt %)</td>
<td>50.5</td>
</tr>
</tbody>
</table>

Figure 3. Cumulative particle size distributions of minerals in coals A and B.

Calcite and kaolinite were selected, blended, and heated in mineral
particle heating experiments to elucidate the effect of mineral
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sample was the same as that for coal B. All of the samples, coals and
minerals, were ground and sieved to 45−100 μm in particle size.
carried out in a lab-scale DTF, whose schematic diagram is shown in Figure 5. The furnace has a length of 2000 mm and an inner diameter of 56 mm. Because the lowest melting point of the CaO−Al2O3−SiO2 ternary system is about 1443 K,21 the two temperatures, 1573 and 1373 K, were selected to study the PM formation with and without significant melting-phase generation. A Sankyo Piotech micro feeder (model MFEV-10) was used to feed samples. Considering the precision of the sample feeder and burn out of used coals, a feeding rate of 0.3 g/min was selected. A simulated air atmosphere was achieved using 21% O2, with N2 being the balance. The gas was supplied in two flows: primary gas and secondary gas, and the volume ratio was 1:3. The particle residence time in the furnace was about 2 s, which was similar to that in actual boilers.

PM was isokinetically collected by a Dekati cyclone (model SAC-65) and a Dekati low-pressure impactor (DLPI) through a water-cooled N2-quenched probe. The normal flow of the vacuum pump was about 10 L/min. The large particles with an aerodynamic diameter of >10 μm were mainly collected by the cyclone, and smaller particles were size-segregated by the DLPI into 13 fractions. The inner diameter of the sampling probe is about 16 mm. The top of the probe is conical, and quenched N2 was injected into the probe there. Through altering the flow rate of quenched N2, the volume ratio of flue gas entering the sampling probe to the flue gas in the furnace was equal to the area ratio of the inlet opening of the sampling probe to the cross-section of the furnace. In this way, isokinetic sampling was achieved. Quenched N2 could dilute the flue gas and suppress the subsequent reaction. The bulk ashes were the total ashes collected by fiber filters without using the cyclone and DLPI. The collection process used in these experiments was developed and has been verified in a series of publications by our group.22−24

The particles collected on each DLPI stage were weighed, and the PM concentration was defined as the mass ratio of particles on the stage to total ash entering the sampling probe. The elemental compositions of the particles were quantified by XRF, and the considered elements were Na, Mg, Al, Si, P, S, K, Ca, and Fe. The central-mode PM was identified as the particles in which the mass fraction of Al clearly increased with an increasing particle size (as shown below). The bulk ashes collected in the mineral particle heating experiments were further characterized by X-ray diffraction (XRD) and scanning electron microscopy equipped with an energy-dispersive spectrometer (SEM−EDS).

3. RESULTS AND DISCUSSION

3.1. Central-Mode PM Formation in Coal Combustion Experiments. The elemental composition of different PM formation modes is quite different.22,25,26 On the basis of this characteristic, Yu et al.27,28 developed a method to identify the PM formation modes using the mass fraction size distribution (MFSD) of aluminum (Al). Figure 6 shows the MFSDs of Al generated in combustion of coals A and B. Similar to Yu et al.,27,28 the distribution profiles are both S-shaped and can be divided into three sections. In the size range of <0.2 μm, the
mass fractions of Al are very low and almost independent of the particle size. In the size range of >2 μm, the mass fractions of Al are the highest and also almost independent of the particle size. In contrast, in the size range of 0.2–2 μm, the mass fractions of Al are between those in the other two size ranges and increase with an increasing particle size. According to the method by Yu et al., three PM formation modes can be identified: ultrafine-mode PM smaller than 0.2 μm (DLPI stages 1–3), central-mode PM between 0.2 and 2 μm (DLPI stages 4–8), and coarse-mode PM larger than 2 μm.

As mentioned above, ultrafine-mode PM is mainly produced by the vaporization–condensation mechanism. Therefore, its elemental composition is closely associated with elemental volatility. Al has a much lower volatility than volatile elements (e.g., Na, K, and S) and other refractory elements (e.g., Si, Ca, and Fe). Therefore, the mass fraction of Al in the ultrafine particles is very low. This has been observed by other investigators. For example, Quann et al. reported that Al2O3 in ultrafine particles accounted for only 0.97%. Linak et al. reported that the Al content in ultrafine particles was notably less than that in larger particles. Volatile elements and Ca and Fe were the main elements in ultrafine-mode PM. On the other hand, because the element vapors are well-mixed, the compositions of condensed fine particles are nearly uniform. Therefore, the mass fraction of Al in ultrafine particles is quite low and nearly independent of the particle size. Coalescence of melting particles contributes greatly to coarse-mode PM formation, and hence, the coarse-mode particles usually have smooth surfaces and small specific surface areas, which lead to less surface condensation of volatile species. The elemental composition of coarse-mode PM is similar to that of bulk coal ash, with the Al content being quite high and almost independent of the particle size. In comparison to coarse-mode particles, the central-mode particles are primarily irregular in shape and have larger specific surface areas. Therefore, more volatile species heterogeneously condense on their surfaces. According to the characteristics of heterogeneous condensation, the amount of volatile elements decreases, whereas the content of refractory elements (including Al) increases with an increasing particle size in central-mode PM. These features have been extensively discussed and verified in our previous work.

The elemental compositions of central-mode PM identified by MFSDs of Al in the coal combustion experiments are shown in Figure 7. Na, K, S, and P are the main volatile elements, and Mg, Ca, Fe, Si, and Al are the main refractory elements. With an increasing particle size, the mass fractions of refractory elements increase, whereas those of volatile elements decrease, which is consistent with other reports. This further confirms the validation of identification of PM formation mode using MFSD of Al in this work.

On the basis of Figure 6, particles between 0.2 and 2 μm (DLPI stages 4–8) are identified as central-mode PM. The concentration of central-mode PM is represented as the mass ratio of the central-mode particles to the total ash entering the sampling probe (mg/g_sub). The results obtained during the combustion of different coals at 1373 and 1573 K are shown in Figure 8. The central-mode PM concentrations for coal A at 1373 and 1573 K are 4.27 and 6.91 mg/g_sub, respectively, while those for coal B are 2.81 and 2.44 mg/g_sub respectively. Clearly, the central-mode PM concentrations for coal A are greater than those for coal B at both temperatures. The variation tendencies in the central-mode PM concentration for the two coals with an increase in the temperature are opposite: the concentration for coal A increases by 61.8%, whereas that for coal B decreases by 13.2%. These results are explained as below.

3.2. Role of Coal Fragmentation and Mineral Coalescence in Central-Mode PM Formation. Coal fragmentation and mineral coalescence play important roles in central-mode PM formation during coal combustion. In the process of fragmentation, a coarse particle fragments into some smaller pieces, which promotes central-mode PM formation. In contrast, when coalescence happens, small particles can aggregate to form coarser particles, which restrains central-mode PM formation. Xu et al. summarized the relationship among coal fragmentation, mineral coalescence, and residual
ash particle size. The result showed that more violent coal fragmentation correlated with weaker mineral coalescence, smaller residual ash particle size, and formation of more central-mode PM.

Coal fragmentation includes char fragmentation and mineral fragmentation. Char fragmentation is influenced by char macroporosity, particle size, and combustion conditions.3,9,32–34 Among them, char macroporosity can be approximately estimated from the maceral composition, specifically, the content of vitrinite in coal.18 Mineral fragmentation is influenced by mineral composition, particle size, and combustion conditions.11,12,35 It is believed that carbonates and pyrite in coal would experience violent fragmentation during combustion and clay minerals would not experience violent fragmentation during combustion.

Mineral coalescence is influenced by melting-phase generation, mineral spatial distribution, and combustion conditions.31,36 Si, Al, Mg, Ca, and Fe are the major ash-forming elements in coal. Experimental studies and theoretical calculations have already shown that Mg, Ca, and Fe species can react with aluminosilicates to form low-temperature eutectics.37–39 The melting point of coal ash was associated with the ratio SiO$_2$/Al$_2$O$_3$ and the content of basic oxides (including Fe$_2$O$_3$ and CaO).37,38 With a constant SiO$_2$/Al$_2$O$_3$ value, the melting-phase generation will first be enhanced and then diminished with increasing basic oxide (e.g., CaO) contents.39 As a result, an optimal ratio of Mg, Ca, and Fe species to aluminosilicates for the maximum melting-phase generation may exist. On the basis of these facts, a factor $Q_a$ is defined as

$$ Q_a = \frac{(\text{MgO} + \text{CaO} + \text{Fe}_2\text{O}_3)}{(\text{MgO} + \text{CaO} + \text{Fe}_2\text{O}_3 + \text{SiO}_2 + \text{Al}_2\text{O}_3)} $$

A summary of related literature data39–46 (see Table 2) indicates that, under typical conditions of coal combustion (air atmosphere, $T = 1373$–$1723\text{ K}$), mineral composition satisfying SiO$_2$/Al$_2$O$_3 = 1$–3 and $Q_a \approx 0.25$ usually led to the minimum central-mode PM/PM$_{2.5}$ formation. If the mineral composition of the raw coal is beyond these criteria, adjustment of the composition by mineral addition to meet them can reduce central-mode PM/PM$_{2.5}$ formation. If the mineral composition of the raw coal conforms to these criteria, further mineral addition would have little influence on central-mode PM/PM$_{2.5}$ reduction. Ninomiya et al.41 concluded that melting-phase generation played a significant role in PM reduction. Therefore, the criteria, i.e., SiO$_2$/Al$_2$O$_3 = 1$–3 and $Q_a \approx 0.25$, may suggest the optimal mineral composition for the maximum melting-phase generation and coalescence occurrence during coal combustion.

These optimal ranges are supported by other studies. Helble et al.10 found that Kentucky no. 11 coal ($Q_a = 0.28$) and Illinois no. 6 coal ($Q_a = 0.29$), whose mineral compositions are close to the optimal criterion $Q_a \approx 0.25$, experienced clear coalescence during combustion. Akiyama et al.47 found that the addition of MgO to three different coals ($Q_a = 0.26, 0.39$, and 0.43) shifted the mineral compositions away from this criterion and led to a reduction of the molten-slag fraction in the resulting ash. Both coal fragmentation and mineral coalescence are influenced by the reaction temperature.31 With an increase in the temperature, the external thermal shock of the particle and the internal gas evolution are both intensified. Therefore, fragmentation is expected to become more severe, which promotes the formation of central-mode PM. On the other hand, the generation of more melting phases at higher temperatures results in more coalescence, which may restrain the formation of central-mode PM. Consequently, the variation of the central-mode PM concentration with an increase in the temperature depends upon the competition between fragmentation and coalescence processes. Many studies3,9,32–34 report that fine PM concentrations increase with an increase in the temperature, which indicates that the extent of fragmentation usually increases more significantly than that of mineral coalescence with an increase in the temperature under the conditions investigated. The above points are illustrated schematically in Figure 9.

### Table 2. Effect of Mineral Addition on Central-Mode PM/PM$_{2.5}$ Control

<table>
<thead>
<tr>
<th>investigation</th>
<th>SiO$_2$/Al$_2$O$_3$</th>
<th>$Q_a^a$</th>
<th>addition minerals</th>
<th>variation of central-mode PM/PM$_{2.5}$ concentration</th>
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<tbody>
<tr>
<td>Ninomiya et al.40</td>
<td>2.75</td>
<td>0.10</td>
<td>Ca/Mg mineral</td>
<td>PM$_{2.5}$ decreased by 15.9% (Ca)/65.5% (Mg)</td>
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<tr>
<td>Wei et al.42</td>
<td>1.71</td>
<td>0.21</td>
<td>Mg minerals</td>
<td>PM$_{2.5}$ decreased by 77.6%</td>
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<tr>
<td>Ninomiya et al.41</td>
<td>2.65</td>
<td>0.05</td>
<td>Mg mineral</td>
<td>PM$_{2.5}$ decreased by 97.5%</td>
</tr>
<tr>
<td>Ji et al.44</td>
<td>1.50</td>
<td>0.25</td>
<td>Si–Al minerals</td>
<td>PM$_{2.5}$ increased</td>
</tr>
<tr>
<td>Wang et al.39</td>
<td>1.60</td>
<td>0.28</td>
<td>Si–Al minerals</td>
<td>obvious decrease</td>
</tr>
<tr>
<td>Zhou et al.45</td>
<td>1.42</td>
<td>0.55</td>
<td>Si–Al minerals</td>
<td>obvious decrease</td>
</tr>
<tr>
<td>Xu et al.46</td>
<td>0.80</td>
<td>0.77</td>
<td>Si–Al minerals</td>
<td>obvious decrease</td>
</tr>
</tbody>
</table>

$^a Q_a = \frac{(\text{MgO} + \text{CaO} + \text{Fe}_2\text{O}_3)}{(\text{MgO} + \text{CaO} + \text{Fe}_2\text{O}_3 + \text{SiO}_2 + \text{Al}_2\text{O}_3)}$
In this study, the combustion parameters for coals A and B were the same. The coal particle size (45–100 μm), vitrinite content (see Table 1), fine mineral particle size (<5 μm) (see Figure 3), and mineral spatial distribution (see Figure 4) in the two coals were all similar. Therefore, the inverse variation tendencies of the central-mode PM concentration with an increase in the temperature were attributed to the different mineral compositions of the two coals. There was more frangible minerals, such as calcite, in coal A than in coal B (17.8% in coal A and 8.8% in coal B). Therefore, the extent of fragmentation of coal A was higher than that of coal B. Meanwhile, the mineral composition of coal A (SiO₂/Al₂O₃ = 2.2; Qₐ = 0.42) was far away from the optimal criteria for melting-phase generation, whereas the composition of coal B (SiO₂/Al₂O₃ = 1.7; Qₐ = 0.22) was closer to the criteria. Consequently, the extent of melting-phase generation for coal B may be higher than that for coal A. For these reasons, the central-mode PM concentration for coal A is expected to be higher than that for coal B at the same temperature, which is consistent with Figure 8. With an increase in the temperature, the extent of fragmentation for coal A would be significantly higher than that of coalescence. This leads to an increase in the central-mode PM concentration. In contrast, for coal B, considering the restraint of fragmentation by plenty melting-phase generation, the extent of coalescence may be higher than that of fragmentation, which results in a decrease in the central-mode PM concentration. These are clearly illustrated in Figure 8.

Because of the complexity of coal combustion, mineral particle heating experiments were also carried out to study the interactions between Mg/Ca/Fe minerals (calcite as a representative) and Si–Al minerals (kaolinite as a representative), which were used to verify their effects on central-mode particle formation at high temperatures.

### 3.3. Mineral Interactions and Central-Mode Particle Formation in Mineral Particle Heating Experiments

The mineral compositions of the bulk ashes generated from heating of calcite + kaolinite at 1373 and 1573 K were obtained by XRD analysis. The results are shown in Figure 10. Anorthite, gehlenite, mullite, portlandite, quartz, and tridymite were detected in both ashes. The appearance of Ca aluminosilicates, anorthite (CaO·Al₂O₃·2SiO₂) and gehlenite (2CaO·Al₂O₃·SiO₂), suggests the interactions between calcite and kaolinite.

![Figure 10. Mineral compositions of calcite + kaolinite bulk ashes generated at 1373 and 1573 K (A, anorthite; G, gehlenite; M, mullite; P, portlandite; Q, quartz; and T, tridymite).](image)

According to the literature reported, calcite and kaolinite are expected to be subjected to the following reactions 1–7 in the mineral particle heating experiments:

1. \[ \text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2 \]
2. \[ \text{Al}_2\text{O}_3\cdot2\text{SiO}_2\cdot2\text{H}_2\text{O} \rightarrow \text{Al}_2\text{O}_3\cdot2\text{SiO}_2 + 2\text{H}_2\text{O} \]
3. \[ 2(\text{Al}_2\text{O}_3\cdot2\text{SiO}_2) \rightarrow 2\text{Al}_2\text{O}_3\cdot3\text{SiO}_2 + \text{SiO}_2 \]
4. \[ 2\text{Al}_2\text{O}_3\cdot3\text{SiO}_2 \rightarrow 2(\text{Al}_2\text{O}_3\cdot2\text{SiO}_2) + \text{SiO}_2 \]
5. \[ 3(\text{Al}_2\text{O}_3\cdot2\text{SiO}_2) \rightarrow 3\text{Al}_2\text{O}_3\cdot2\text{SiO}_2 \text{ (mullite) + SiO}_2 \]
6. \[ 2\text{Al}_2\text{O}_3\cdot3\text{SiO}_2 + \text{CaO} \rightarrow \text{CaO}·\text{Al}_2\text{O}_3·2\text{SiO}_2 \text{ (anorthite) + Al}_2\text{O}_3·\text{SiO}_2 \]
7. \[ 2\text{Al}_2\text{O}_3·3\text{SiO}_2 + 4\text{CaO} \rightarrow 2(2\text{CaO}·\text{Al}_2\text{O}_3·\text{SiO}_2) \text{ (gehlenite) + SiO}_2 \]

When various Ca aluminosilicates are generated, eutectics with low melting points between 1373 and 1573 K may appear. The morphologies and compositions of the calcite + kaolinite bulk ashes generated at 1373 and 1573 K are shown in Figure 11. There are two kinds of typical particles in the 1373 K ash (see Figure 11a). The particles with rough surfaces contain more Ca but less Si and Al. In contrast, the particles with smooth surfaces contain more Si and Al but less Ca. These

![Figure 11. Morphologies and local compositions of calcite + kaolinite bulk ashes generated at different temperatures.](image)
rough and smooth particles are produced by calcite and kaolinite, respectively. No obvious connections can be found between these particles. In the 1573 K ash (see Figure 11b), there are also two kinds of particles. Unlike the ash particles generated at 1373 K, there are clear connections between the particles generated at 1573 K, and these connections contain large amounts of Si, Al, and Ca. These observations suggest the generation of the melting phase and the occurrence of coalescence at 1573 K.

As illustrated in Figure 9, both particle fragmentation and coalescence that resulted from melting-phase generation affect the formation of fine particles. This can be further demonstrated by correlating fine particle formation with the generation of melting phases. The melting phases generated during heating of calcite, kaolinite, and calcite + kaolinite were calculated using FactSage 5.4. The detailed information on the calculation was described in section 2.3. The calculation results and the fine particle (<10 μm) concentrations generated during heating of calcite, kaolinite, and calcite + kaolinite at 1373 and 1573 K are shown in Figure 12. With an increase in the temperature, the variations of the simulated melting-phase formation at 1373 K are shown in Figure 12. With an increase in the temperature e... | 1.64 | 5.87 | 14.5, and 5.87 | m) concentrations generated during heating of calcite, kaolinite, and calcite + kaolinite at 1373 and 1573 K are shown in Figure 12. With an increase in the temperature e... | 1.64 | 5.87 | 14.5, and 5.87 | m) concentrations generated during heating of calcite, kaolinite, and calcite + kaolinite at 1373 and 1573 K are shown in Figure 12. With an increase in the temperature e...

4. CONCLUSION
Coal combustion and mineral particle heating experiments were carried out in a DTF at 1373 and 1573 K to study the temperature effect on central-mode PM formation based on mineral composition. The major conclusions are as follows: (1) The mass fraction size distribution of Al can be used to effectively identify the different formation modes of PM generated during coal combustion. (2) Under typical conditions of coal combustion (air atmosphere, T = 1373–1723 K), the mineral composition satisfying SiO2/Al2O3 = 1–3 and Qf = (MgO + CaO + Fe2O3)/(MgO + CaO + Fe2O3 + SiO2 + Al2O3) ≈ 0.25 is optimal for the generation of melting phases and the occurrence of coalescence. With an increase in the temperature, central-mode PM formation usually increases for coals with a mineral composition beyond these criteria. In contrast, central-mode PM formation does not increase or even decrease for coals with a mineral composition close to these criteria. (3) Interactions between Mg/Ca/Fe minerals and Si–Al minerals are observed. They influence particle fragmentation and coalescence and, thus, influence the variation tendency of central-mode PM formation with an increase in the temperature.

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Notes
The authors declare no competing financial interest.

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