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# Improving the efficiency of CaO-based sorbent by SrCO<sub>3</sub> for high-temperature sulfur removal during coal combustion

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**Abstract:** With the goal of adding the appropriate amount of SrCO<sub>3</sub> to coal, the synergistic effects of SrCO<sub>3</sub> and desulfurization agents were studied by using a sulfur determination device and a carbon–sulfur spectrometer. Mineral constituents were investigated with an XRD diffraction analyzer. Results show that, at a given temperature, desulfurization efficiency increased in varying degrees based on the proportion of added SrCO<sub>3</sub>. An increasing amount of calcium sulphoaluminates also appeared in the ash residue. Meanwhile, SrCO<sub>3</sub> sulfur capture capacity improved when the temperature reached 1200°C because of the large amount of generated calcium sulphoaluminates (3CaO·3Al<sub>2</sub>O<sub>3</sub>·CaSO<sub>4</sub>) and the controlled decomposition of CaSO<sub>4</sub>. At temperatures of 1200°C–1300°C, the ascending rate of the sulfur-fixing ratio decreased due to the sharp decomposition of CaSO<sub>4</sub> and the pyrolysis of a small amount of calcium sulphoaluminate. From 1300°C, the decomposition of increasing amounts of calcium sulphoaluminate gradually worsened the desulfurization. The diffraction peak of calcium sulphoaluminate was the strongest at 1300°C and the sulfur-fixing ratio was also the largest. Therefore, calcium sulphoaluminate reached the highest level at this temperature.

**Keywords:** calcium-based sorbent; calcium sulphoaluminate; efficiency; SrCO<sub>3</sub>.

## 1 Introduction

Coal is an important resource in China. The strong demand for coal is the main driver of coal-fired power plants. However, the combustion of pulverized coal

releases harmful gases (e.g. SO<sub>2</sub>), which form acid rain and damages the environment. Adding calcium-based sorbent to coal significantly improves the sulfur-fixing effect at low temperature. However, the improvement is not ideal at high temperatures, especially over 1200°C. In some studies, such additives as CeO<sub>2</sub> [1], Cr<sub>2</sub>O<sub>3</sub> [1, 2], Fe<sub>2</sub>O<sub>3</sub> [3], MnO<sub>2</sub> [4], Al<sub>2</sub>O<sub>3</sub> [1], SiO<sub>2</sub> [1], and MgO [5, 6], are added to calcium-based sorbent to generate high-temperature stable phases, such as 3CaO·3Al<sub>2</sub>O<sub>3</sub>·CaSO<sub>4</sub> (i.e. C<sub>4</sub>A<sub>3</sub>S) [7, 8], BaSO<sub>4</sub>, and Ca<sub>5</sub>(SiO<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> [9]. Hence, the sulfur-fixing effect is improved by preventing or postponing the decomposition of CaSO<sub>4</sub> (desulfurization product) [10, 11].

Strontium and calcium are both alkaline earth metals. The addition of strontium salt results not only in the generation of SrSO<sub>4</sub>, which is relatively stable at a high temperature, but also in the increased amounts of generated C<sub>4</sub>A<sub>3</sub>S. However, the reaction mechanism and decomposition temperature of C<sub>4</sub>A<sub>3</sub>S varies depending on certain conditions [12–14]. On the basis of Ca/S = 2, Guozhen Lin et al. added silicon, iron, and strontium to coal and found that, after high-temperature combustion (more than 1200°C), the main phases in the Sr system are SrSO<sub>4</sub> and C<sub>4</sub>A<sub>3</sub>S, whereas in the Si and Fe system, the main phase is CaSO<sub>4</sub> [15]. By analyzing the sulfur-fixing principles of different sulfur adsorbents at high temperature, Peng Bai found that the sulfur-fixing efficiencies of barium salt and strontium salt with an equal mole ratio (M/S = 1 and 2) are higher than those of calcium-based sulfur adsorbents [16]. However, their study did not analyze the mineral composition of coal ash. Hong Zhang discovered that strontium carbonate can improve the desulfurization efficiency of limestone [17] and that SrSO<sub>4</sub> can be found in coal ash [18]. In other studies, pure chemical reagents, such as CaO, Al<sub>2</sub>O<sub>3</sub>, and CaSO<sub>4</sub>, are mixed according to certain proportions to study the generation mechanism of C<sub>4</sub>A<sub>3</sub>S [19–23].

Based on the low utilization rate of calcium and the poor effect of high-temperature sulfur fixation in the actual production process, the synergistic effects of SrCO<sub>3</sub> and desulfurization agents on the formation of C<sub>4</sub>A<sub>3</sub>S, mineral constituents, and sulfur-fixing efficiency are investigated by employing a sulfur determination

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device, an XRD diffraction analyzer, and so on. This process is advantageous in enhancing the sulfur-fixing efficiency and increasing the use of high-sulfur coal.

## 2 Materials and methods

### 2.1 Reagents and materials

The pulverized coal used in this experiment was high-sulfur coal (41.92%  $FC_d$ ; 9.04%  $V_d$ ; 49.04%  $A_d$ ; 7.02%  $S_{t,d}$ ). Compositions of Ash are shown in Table 1. The following materials (pure chemical reagent) were prepared:  $Ca(OH)_2$ , (Jinbei Fine Chemical Co. Ltd., Tianjin, China) as sulfur-fixing agent and  $SrCO_3$ , (Zhiyuan Chemical Reagent Co. Ltd., Tianjin, China) as additive. Ca/S mole ratio was equal to 1.5.

The high-sulfur coal and  $Ca(OH)_2$  were taken and mixed according to a given proportion. About 1%, 3%, and 5%  $SrCO_3$  were added to coal samples, respectively. They were then blended and ground in a grinding bowl to prepare the samples. A certain amount of coal, which was taken from the samples, was placed in the corundum crucible, and then calcined at 1200°C, 1300°C, and 1400°C and preserve heat for 1 h, respectively. The ash residues were collected to determine the sulfur content.

**Table 1:** Compositions of ash in high-sulfur coal.

Compositions	$SiO_2$	$Fe_2O_3$	$Al_2O_3$	CaO	MgO	$K_2O$	$Na_2O$
Contents (%)	51.76	20.19	21.46	2.24	1.12	1.96	1.27

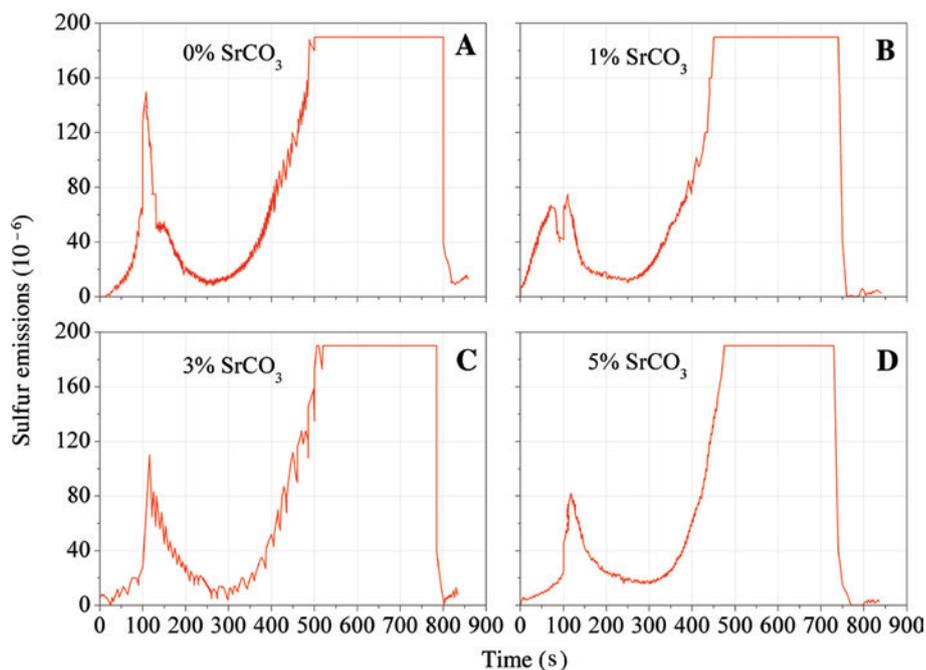
### 2.2 Experimental equipment

In the current work, a YX-DL8300 Sulphur Determination Device (Changsha Youxin Instrument Manufacturing Co. Ltd., Changsha, China) was used to verify the sulfur content in the coal samples. An ash melting point detector (YX-HRD3000, Changsha Qiulong Equipment Co. Ltd., Changsha, China) was utilized to calcine the coal samples, after which an infrared carbon-sulfur spectrometer (ZY-HWC8, Nanjing Zhengyuan Analytical Instrument Manufacturing Co. Ltd., Nanjing, China) was used to ascertain the sulfur content in the ash. The changes of the mineral constituents in coal ash were investigated through an XRD diffraction analyzer (DX-2700, Dandong Fangyuan Instrument Co. Ltd., Dandong, China).

## 3 Results and discussion

### 3.1 Effects of additive proportion on high sulfur coal desulfurization

The sulfur-releasing rates of the coal samples with different amounts of  $SrCO_3$  when temperatures reach 1100°C are shown in Figure 1. Although the size, shape, and location of each peak vary, the changing trends are basically the same, that is, the curves evidently present twin peaks. The two peaks indicate the release process of organic sulfur (i.e. aliphatic, aromatic, etc.) and inorganic sulfur (i.e. sulfate, iron sulfide, etc.). The decomposition temperature of inorganic sulfur is comparatively high, and the decomposition



**Figure 1:** Rules for sulfur release during the combustion of coal samples.

rate is lower than that of organic sulfur (i.e. with a lower decomposition temperature); thus, the second peak is flat. After 300 s, the released sulfur concentration begins to rise rapidly. Then, it is maintained for approximately 300 s, when it approaches 200 ppm. Finally, sulfur in the coal sample is released completely. A large transition interval exists between two peaks as a result of the decomposition of pyrite (around 600°C) to sulfur [24–27].

Compared with the case in which no strontium carbonate is added, the first peak is pointed, and the peak value is higher, indicating that more sulfur is released in a relatively short time. After the addition of 1% strontium carbonate, sulfur precipitation accelerates, and the peak appears in advance, but the peak value decreases obviously; further, when the addition ratio is 3% or 5%, the presence of the peak is delayed. It is possible that, to a certain extent, a small amount of strontium carbonate (1% SrCO<sub>3</sub>) plays a catalytic role in the release of sulfur, although this assumption requires further research). However, when more strontium carbonate (3% or 5% SrCO<sub>3</sub>) is added, the actions of desulphurization begin to hold a dominant position.

With the increase in SrCO<sub>3</sub> content, the first peak value decreases and the duration of the second peak reduces, suggesting that at 1100°C, the addition of 3% or 5% SrCO<sub>3</sub> promotes sulfur fixation.

### 3.2 Influence of temperature on the desulfurization effect

Desulfurization rate ( $\eta_s$ ) evaluates the sulfur fixation effect and is calculated on the basis of the formula expressed as

$$\eta_s = \frac{S_0 - S_1}{S_0} \quad (1)$$

where  $S_0$  is the initial sulfur content in coal sample (7.02%), and  $S_1$  is the sulfur content that escaped in the form of gas during combustion under different conditions.

Desulfurization rates are plotted in Figure 2 as a function of the temperature from 1100°C to 1400°C. The changing trends of the curves are roughly the same, that is, the desulfurization rate increases first and then descends. After adding SrCO<sub>3</sub>, the desulfurization rate enhances greatly (the higher the temperature, the more obvious the effect of SrCO<sub>3</sub>). It is worth mentioning that, after adding 3% SrCO<sub>3</sub> to the coal samples, the sulfur-fixing ratio slightly increases compared with the case in which 1% SrCO<sub>3</sub> is added at 1100°C or 1400°C. Meanwhile, at 1200°C or 1300°C, the increase is larger, but the desulfurization rate is slightly less than the value of the coal sample with

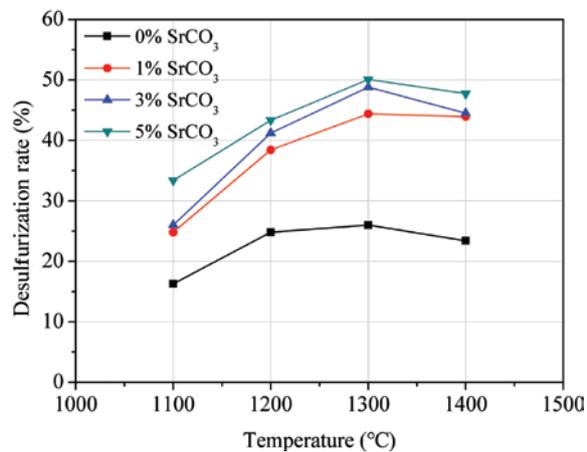


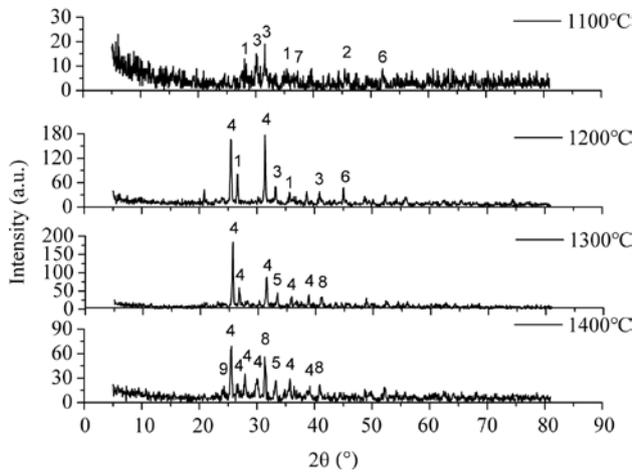
Figure 2: Effects of temperature on sulfur fixation.

5% SrCO<sub>3</sub>. This finding implies that at 1200°C, some compounds that keep the desulfurization product (e.g. CaSO<sub>4</sub>) from decomposing are generated. Otherwise, this finding could also mean that CaSO<sub>4</sub> further reacts to form new sulfur retention phases that are stable at high temperature, which in turn, induces a sulfur-fixing ratio that increases significantly. When the temperature is higher than 1300°C, the sulfur-fixing ratio shows a declining trend. This may be due to the weakening effect of the inhibition of desulfurization product decomposition or the disintegration of the decomposition of sulfur retention phases at this temperature. When temperature exceeds a certain value, sulfur retention phases decompose gradually and release sulfur again. Previous studies have shown that this compound is C<sub>4</sub>A<sub>3</sub>S [28].

### 3.3 Influence of temperature on the formation of calcium sulphoaluminate

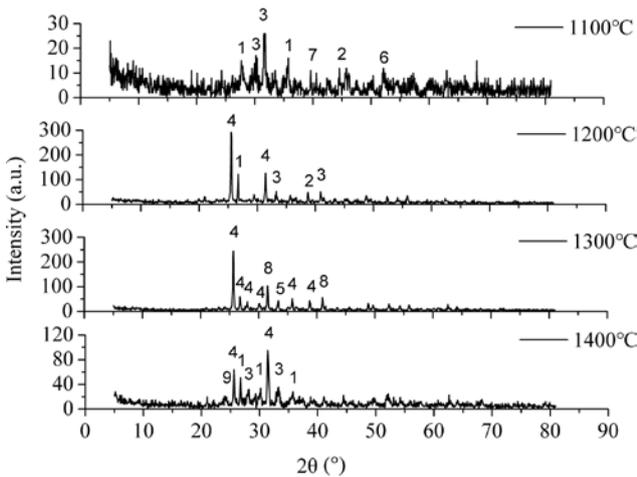
The XRD spectra of different coal ash samples at different temperatures are presented in Figures 3 and 4. As can be seen in both figures, no characteristic peak of C<sub>4</sub>A<sub>3</sub>S appears at 1100°C, and the main mineral is 3CaO·5Al<sub>2</sub>O<sub>3</sub>. In addition, small amounts of CaO·2Al<sub>2</sub>O<sub>3</sub>, 12CaO·7Al<sub>2</sub>O<sub>3</sub> (in Figure 4), and residual CaSO<sub>4</sub>, Ca(OH)<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> are observed. These observations indicate that the formation temperature of 3CaO·5Al<sub>2</sub>O<sub>3</sub> is lower than that of C<sub>4</sub>A<sub>3</sub>S and that its generation speed is faster than that of C<sub>4</sub>A<sub>3</sub>S.

As shown in Figures 3 and 4, when the temperature rises to 1200°C, C<sub>4</sub>A<sub>3</sub>S appears in the mineral phases, and CaSO<sub>4</sub>, Ca(OH)<sub>2</sub> (in Figure 4), and 3CaO·5Al<sub>2</sub>O<sub>3</sub> still exist simultaneously, suggesting that, within the temperature range, only a part of CaSO<sub>4</sub> and CaO react with Al<sub>2</sub>O<sub>3</sub> to



**Figure 3:** XRD spectra of coal ash samples at different temperatures (3% SrCO<sub>3</sub>).

(1) CaSO<sub>4</sub>, (2) Ca(OH)<sub>2</sub>, (3) Ca<sub>3</sub>Al<sub>10</sub>O<sub>18</sub>, (4) 3CaO·3Al<sub>2</sub>O<sub>3</sub>·CaSO<sub>4</sub>, (5) Ca<sub>12</sub>Al<sub>14</sub>O<sub>33</sub>, (6) Al<sub>2</sub>O<sub>3</sub>, (7) CaAl<sub>4</sub>O<sub>7</sub>, (8) Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>, and (9) SrSO<sub>4</sub>.



**Figure 4:** XRD spectra of coal ash samples at different temperatures (5% SrCO<sub>3</sub>).

(1) CaSO<sub>4</sub>, (2) Ca(OH)<sub>2</sub>, (3) Ca<sub>3</sub>Al<sub>10</sub>O<sub>18</sub>, (4) 3CaO·3Al<sub>2</sub>O<sub>3</sub>·CaSO<sub>4</sub>, (5) Ca<sub>12</sub>Al<sub>14</sub>O<sub>33</sub>, (6) Al<sub>2</sub>O<sub>3</sub>, (7) CaAl<sub>4</sub>O<sub>7</sub>, (8) Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>, and (9) SrSO<sub>4</sub>.

form high-temperature, stable C<sub>4</sub>A<sub>3</sub>S. In addition, CaSO<sub>4</sub> still exists at this temperature, indicating that the addition of SrCO<sub>3</sub> may melt and harden on the surface of the sulfur fixation agent, wrap the CaSO<sub>4</sub>, and prevent CaSO<sub>4</sub> from decomposing to a certain extent.

Furthermore, the figures show that, the C<sub>4</sub>A<sub>3</sub>S crystals grow larger at 1300°C and that the diffraction peak intensity continues to strengthen, but the growth slows down, and decomposition occurs. At this point, 3CaO·5Al<sub>2</sub>O<sub>3</sub>, CaSO<sub>4</sub>, and Ca(OH)<sub>2</sub> have all disappeared, and only small amounts of 3CaO·Al<sub>2</sub>O<sub>3</sub> and 12CaO·7Al<sub>2</sub>O<sub>3</sub> appear. This may be attributed to the following series of reactions: CaO first reacts with

Al<sub>2</sub>O<sub>3</sub> to generate CaO·2Al<sub>2</sub>O<sub>3</sub>, and then CaO·2Al<sub>2</sub>O<sub>3</sub> reacts with CaO, CaSO<sub>4</sub>, and Al<sub>2</sub>O<sub>3</sub> to form C<sub>4</sub>A<sub>3</sub>S and 12CaO·7Al<sub>2</sub>O<sub>3</sub>; then, 3CaO·Al<sub>2</sub>O<sub>3</sub> is generated by the decomposition of C<sub>4</sub>A<sub>3</sub>S at a high temperature. This finding indicates that a small amount of C<sub>4</sub>A<sub>3</sub>S has already decomposed at 1300°C.

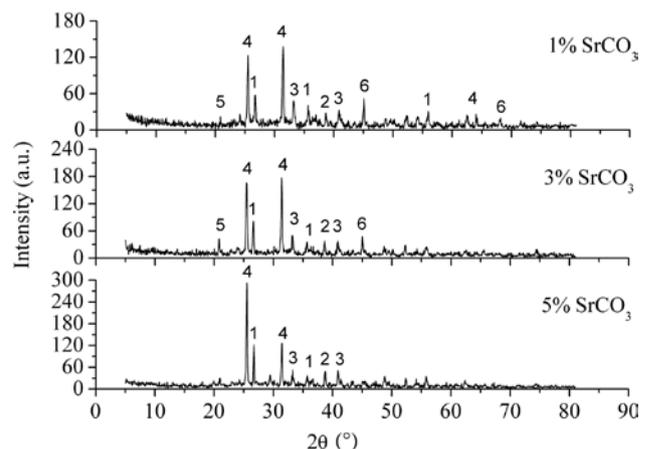
As temperature continues to rise to 1400°C, SrSO<sub>4</sub> appears in the ash residue in Figures 3 and 4. However, the peak intensity of C<sub>4</sub>A<sub>3</sub>S starts to decline rapidly. In the coal ash sample with 3% SrCO<sub>3</sub> (Figure 3), the peak intensity of 3CaO·Al<sub>2</sub>O<sub>3</sub> increases, whereas in the coal ash sample with 5% SrCO<sub>3</sub> (Figure 4), CaSO<sub>4</sub> and 3CaO·Al<sub>2</sub>O<sub>3</sub> reappear, suggesting that the significant pyrolysis of C<sub>4</sub>A<sub>3</sub>S occurs at 1400°C.

In summary, adding a certain amount of SrCO<sub>3</sub> (3% SrCO<sub>3</sub> or 5% SrCO<sub>3</sub>) to coal promotes the formation of C<sub>4</sub>A<sub>3</sub>S. Under this experimental condition, C<sub>4</sub>A<sub>3</sub>S is generated rapidly at around 1200°C. At 1300°C, the reaction continues, and decomposition begins to occur. At 1400°C, decomposition obviously increases. Therefore, the best calcining temperature for the synthesis of C<sub>4</sub>A<sub>3</sub>S is between 1200°C and 1300°C.

### 3.4 Effects of additive proportion on the formation of calcium sulphoaluminate

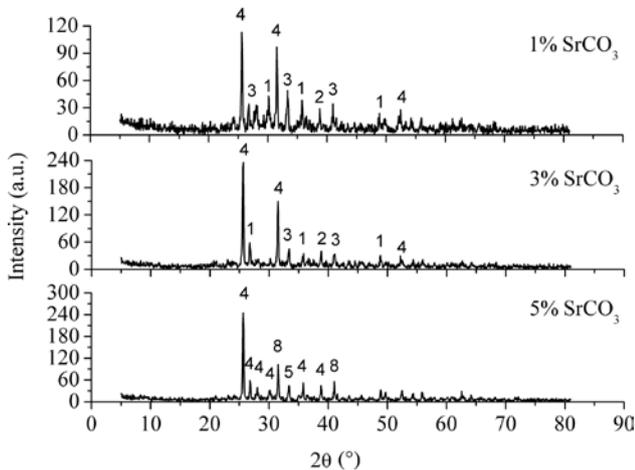
According to the above analysis, although the sulfur retention product CaSO<sub>4</sub> decomposes at high temperature, the existence of high temperature phase C<sub>4</sub>A<sub>3</sub>S increases the sulfur-fixing rate owing to the addition of SrCO<sub>3</sub>.

To determine the appropriate proportion of SrCO<sub>3</sub>, the mineral constituents of the coal ash samples were analyzed at 1200°C and 1300°C, as shown in Figures 5 and 6,



**Figure 5:** XRD spectra of coal ash samples at 1200°C.

(1) CaSO<sub>4</sub>, (2) Ca(OH)<sub>2</sub>, (3) Ca<sub>3</sub>Al<sub>10</sub>O<sub>18</sub>, (4) 3CaO·3Al<sub>2</sub>O<sub>3</sub>·CaSO<sub>4</sub>, (5) Ca<sub>12</sub>Al<sub>14</sub>O<sub>33</sub>, (6) Al<sub>2</sub>O<sub>3</sub>, (7) CaAl<sub>4</sub>O<sub>7</sub>, and (8) Ca<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>.



**Figure 6:** XRD spectra of coal ash samples at 1300°C. (1)  $\text{CaSO}_4$ , (2)  $\text{Ca}(\text{OH})_2$ , (3)  $\text{Ca}_3\text{Al}_2\text{O}_8$ , (4)  $3\text{CaO}\cdot 3\text{Al}_2\text{O}_3\cdot \text{CaSO}_4$ , (5)  $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ , (6)  $\text{Al}_2\text{O}_3$ , (7)  $\text{CaAl}_2\text{O}_7$ , and (8)  $\text{Ca}_3\text{Al}_2\text{O}_6$ .

respectively. As can be seen, the solid reaction products are basically the same. Given the addition of different quantities of  $\text{SrCO}_3$ , the diffraction peak intensity of the generated phases is also different. With the addition of 1%  $\text{SrCO}_3$  at 1200°C or 1300°C,  $\text{C}_4\text{A}_3\text{S}$  is discovered. With the addition proportions increasing to 3% and 5%, the peak intensity of  $\text{C}_4\text{A}_3\text{S}$  increases because the addition of  $\text{SrCO}_3$  accelerates the sulfide and promotes the creation of  $\text{C}_4\text{A}_3\text{S}$ . However, based on Figure 2, at 1200°C, the increase in the sulfur-fixing ratio with the addition proportion of  $\text{SrCO}_3$  increasing from 1% to 3% is similar to that with the addition proportion increasing from 3% to 5%. At 1300°C, the growth in the sulfur-fixing ratio with the addition proportion of  $\text{SrCO}_3$  increasing from 1% to 3% is higher than that with the proportion increasing from 3% to 5%. Thus, the best addition proportion of  $\text{SrCO}_3$  should be around 3%.

## 4 Conclusions

By adding different proportions of  $\text{SrCO}_3$  to coal samples, the effects of  $\text{SrCO}_3$  on desulfurization efficiency at different temperatures are analyzed and the calcined ashes are examined through XRD. The addition of  $\text{SrCO}_3$  promotes the generation of  $\text{C}_4\text{A}_3\text{S}$  and prevents the pyrolysis of  $\text{CaSO}_4$  at high temperatures to a certain extent, greatly increasing the sulfur-fixing ratio.  $\text{C}_4\text{A}_3\text{S}$  grows rapidly at 1200°C, slows down at 1300°C, and decomposes quickly at 1400°C. At a certain temperature, the sulfur-fixing ratio rises with an increase in the addition proportion of  $\text{SrCO}_3$ . Finally, the sulfur-fixing effect is improved when 3%  $\text{SrCO}_3$  is added at temperatures between 1200°C and 1300°C.

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**Conflict of interest statement:** The authors declare that they have no conflict of interest regarding the publication of this paper.

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