Research Article

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Improvement and prediction on high temperature melting characteristics of coal ash

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Abstract: In order to improve the high temperature melting characteristics of bituminous coal with low ash melting point, three kinds of anthracites were used to improve the ash melting characteristics of blended coal to meet the requirement of blast furnace injection. The complete melting temperature of pulverized coal ash had been calculated by using FactSage thermodynamic calculation software. The results showed that after adding different proportions of anthracite with high ash melting point, the deformation temperature, softening temperature, hemispherical temperature, and flow temperature of the blended coal increased. After adding different proportions of Yang Quan anthracite, compared to Bu Lian Ta bituminous coal, the ash melting point of blended coal increased by 98, 136, 149, and 170 K, respectively. The relationship between the ash melting point of pulverized coal and the calculated value of ash complete melting temperature was obtained as: T_{ST} = 0.7098T_C + 257.98.

Keywords: bituminous coal, anthracite, high temperature melting characteristics, ash fusion point, thermodynamic calculation

1 Introduction

As a kind of fossil energy, coal is one of the most used energy sources [1]. The importance of coal is related to energy security. In 2020, coal accounted for 56.8% of China's primary energy consumption. By 2030, this proportion will drop to about 50% [2]. As the industry with the largest carbon emissions among the 31 manufacturing categories, the steel industry is also facing tremendous pressure and challenges in carbon emissions [3–5]. Blast furnace pulverized coal injection can greatly reduce the consumption of coke in the iron making process [6,7] and lower the cost of iron making [8,9]. Since its popularization in the 1960s, blast furnace coal injection technology has become one of the important ways to save energy and reduce emissions in the steel smelting process and promote the green development of steel enterprises [10–14]. In the current world, especially in China, the utilization of high-quality coal resources is increasing year by year, and reserves are decreasing. How to realize the efficient use of low-rank coal is the key to the sustainable development of energy-consuming industries [15–18].

Low-rank coal is a kind of coal with a low degree of coalification. Shendong bituminous coal has low ash, low sulfur, low phosphorus, and good combustion rate, and is suitable for blast furnace injection [18]. In the practice of blowing Shendong bituminous coal in a large number of blast furnaces, due to its low ash melting point, the ash melting point of blended coal cannot meet the requirements of blast furnace injection (below 1,533 K), and it is easy to cause tuyere slagging [19]. In the process of blast furnace injection, mixed injection of bituminous coal and anthracite is generally used. While reducing fuel costs, mixed injection of bituminous coal and anthracite is also beneficial to increase the combustion rate of injected pulverized coal [20]. In this study, Bu Lian Ta (BLT) bituminous coal with low ash melting point was selected as the research object. Three kinds of anthracites with high ash melting, Yang Quan (YQ), Lu An (LA), and Jiao Zuo (JZ), were added. The ash melting characteristics of different proportions of high ash melting point anthracite to BLT bituminous coal were explored. And coal blending to improve the ash melting characteristics of blended coals was studied. The phase equilibrium calculation function of FactSage thermodynamic calculation software was used to calculate the complete melting temperature during the process of dividing coal ash into group elements to generate liquid phase. The calculation results were linked to the ash melting characteristics. The relationship between the software calculation result and the experimental

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measurement value was explored. The relationship between the value calculated by the software for the complete melting temperature of ash content (liquid phase temperature) and the experimental value of the ash melting characteristic was established. The prediction of coal ash melting characteristic temperature was realized.

2 Materials and methods

2.1 Experimental materials

In this experiment, BLT bituminous coal from Shendong mining area was selected as the research object. The main reason for the low ash melting point is the high content of calcium and iron alkaline oxides, and the Fe_2O_3 and CaO in the ash are as high as 22.92 and 31.70%, respectively. The softening temperature of its coal ash is only 1,503 K, which belongs to the coal type that is easily slagging.

Three kinds of low volatile coals widely used in China were selected and mixed them with BLT bituminous coal. Then, the variation law of melting characteristics of blended coal ash was explored, according to the national standards (GB/T 212-2008 and GB/T 31391-2015) of China. The industrial analysis and elemental analysis of four pulverized coal ash were analyzed, and the results are shown in Table 1. The ash melting characteristics of pulverized coal were measured by the melting point and rate measuring instrument according to the Chinese patent (CN 201910105185.5). The chemical composition of ash was analyzed by X-ray fluorescence. The ash melting characteristics and main chemical composition of the four kinds of pulverized coal ashes are shown in Table 2.

DT is the temperature at which the tip or edge of the ash cone begins to round or bend. ST is the temperature at which the ash cone is bent until the cone touches the pallet or the ash cone becomes spherical. HT is a gray cone to approximate hemispherical shape. FT is the temperature at which the ash cone melts and expands into a thin layer with a height of less than 1.5 mm.

2.2 Research methods

2.2.1 Experimental scheme

The YQ, JZ, and LA anthracite were mixed with BLT bituminous coal. The four ratios of 5:5, 6:4, 7:3, and 8:2 of anthracite to bituminous coal were mixed, respectively. The resulting blending index and ash rate are shown in Table 3.

It can be seen from Table 3 that the volatile fraction content of blended coal was controlled between 11 and 20%, and can meet the actual smelting needs of different blast furnaces. At the same time, under this ratio, the percentage of BLT bituminous coal ash in blended coal ash was evenly distributed between 13 and 45%. The change of ash partial melting characteristics of blended coal with different bituminous coal ash ratios can be studied.

Table 1: Proximate and ultimate analyses of pulverized coal, wt% (dry basis)

Description of sample	Proximate analysis			Ultimate analysis				
	V_d	A_d	FC_d	C	H	N	O	S
BLT	30.00	7.58	62.42	74.59	3.95	0.76	12.64	0.21
YQ	7.49	11.58	80.93	81.20	3.11	1.24	2.09	0.67
JZ	6.85	9.39	83.76	80.59	2.07	0.93	1.82	0.42
LA	11.03	10.43	78.54	83.03	3.55	1.37	2.47	0.34

Table 2: Ash melting temperature and main chemical composition of coal ash[a]

Description of sample	Ash melting temperature (K)				Main chemical composition of coal ash (wt%)					
	DT	ST	HT	FT	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3
BLT	1,465	1,503	1,525	1,538	21.17	10.64	22.92	31.70	0.89	8.72
YQ	>1,673	>1,673	>1,673	>1,673	53.05	35.69	2.55	2.36	0.32	0.70
JZ	>1,673	>1,673	>1,673	>1,673	48.30	33.53	5.71	4.14	0.61	1.15
LA	>1,673	>1,673	>1,673	>1,673	50.08	34.46	4.10	4.07	0.41	1.74

[a] DT, deformation temperature; ST, softening temperature; HT, hemisphere temperature; and FT, flow temperature.

Table 3: Coal blending scheme and proximate analysis

Description of sample	Code	Pulverized coal ratio	Ash constituent of bituminous coal (wt%)	Proximate analysis of blended coal (wt%)		
				V_d	A_d	FC_d
YQ:BLT	A1	50:50	40.12	18.75	9.58	71.67
	A2	60:40	30.56	16.49	9.98	73.53
	A3	70:30	21.87	14.24	10.38	75.38
	A4	80:20	13.79	11.99	10.78	77.23
JZ:BLT	B1	50:50	44.75	18.43	8.48	73.09
	B2	60:40	35.06	16.11	8.67	75.22
	B3	70:30	25.93	13.79	8.85	77.36
	B4	80:20	16.67	11.48	9.03	79.49
LA:BLT	C1	50:50	42.20	20.51	9.01	70.48
	C2	60:40	32.43	18.62	9.29	72.09
	C3	70:30	23.66	16.72	9.58	73.70
	C4	80:20	15.25	14.82	9.86	75.32

2.2.2 Ash melting characteristic experiment

Different proportions of pulverized coal were mixed evenly according to the experimental scheme in Table 3, and then put into a muffle furnace for complete ashing at 1,088 K. The obtained ash samples were tested for ash melting characteristics by the melting point and rate measurement system of Metallurgy Experimental Center of University of Science and Technology Beijing. The temperature of the sample increased from 1,273 to 1,673 K. In this temperature range, the deformation temperature, softening temperature, hemispheric temperature, and flow temperature of ash samples were automatically recorded by the computer.

2.2.3 Calculation of ash melting characteristics

The phase equilibrium calculation was carried out for the ash content of BLT bituminous coal and the oxides in the ash content of the mixed coal sample after being mixed with high ash melting point anthracite. It can be seen from Table 2 that the main oxides in the four kinds of pulverized coal were SiO_2, Al_2O_3, Fe_2O_3, and CaO. The $CaO–SiO_2–Al_2O_3–Fe_2O_3$ quaternary system melt was selected for the calculation. The mass fractions of the four oxides (S_iO_2, Al_2O_3, Fe_2O_3, and CaO) were divided into hundreds. The contents of four main oxides in the ash are shown in Table 4.

The melting process of multicomponent slag system occurred in a specific temperature range. FactSage thermodynamic calculation software can calculate the "Complete melting temperature" of multiple melts. It is the temperature when all solid phases in the system were completely melted into liquid phase. The calculated temperature results correspond to the "Liquidus temperature" in the phase diagram.

The calculation process of "complete melting temperature" of multicomponent melts by FactSage software is shown in the appendix.

3 Results and discussion

3.1 Improvement of ash melting characteristics

The experimental results of ash melting characteristics are shown in Table 5. Three various ratios of anthracite

Table 4: The main oxide content in ash of single coal and blended coal, wt%

Description of sample	SiO_2	Al_2O_3	Fe_2O_3	CaO
BLT	24.49	12.31	26.52	36.68
YQ	56.65	38.11	2.72	2.52
JZ	52.68	36.57	6.23	4.52
LA	54.02	37.17	4.42	4.39
A1	43.75	27.76	12.27	16.22
A2	46.82	30.23	9.99	12.96
A3	49.61	32.47	7.93	9.99
A4	52.21	34.55	6.01	7.23
B1	40.07	25.71	15.31	18.91
B2	42.80	28.07	13.34	15.79
B3	45.37	30.28	11.49	12.86
B4	47.98	32.53	9.61	9.88
C1	41.56	26.68	13.75	18.01
C2	44.44	29.11	11.59	14.86
C3	47.03	31.29	9.65	12.03
C4	49.52	33.38	7.79	9.31

Table 5: Experimental results of ash melting characteristics of different blended coal schemes

Code	Ash content of bituminous coal (wt%)	Ash melting characteristic temperature (K)			
		DT	ST	HT	FT
A1	40.12	1,551	1,601	1,648	1,659
A2	30.56	1,565	1,639	1,664	>1,673
A3	21.87	1,597	1,652	>1,673	>1,673
A4	13.79	1,612	>1,673	>1,673	>1,673
B1	44.75	1,513	1,547	1,598	1,623
B2	35.06	1,545	1,548	1,634	1,655
B3	25.93	1,562	1,593	1,640	1,669
B4	16.67	1,575	1,608	1,650	>1,673
C1	42.20	1,493	1,535	1,653	1,672
C2	32.43	1,529	1,608	1,658	1,672
C3	23.66	1,612	1,641	1,664	>1,673
C4	15.25	1,612	1,654	1,667	>1,673

were mixed with BLT bituminous coal. The ash melting characteristics of blended coal significantly improved compared to the ash melting characteristics of BLT bituminous coal. The minimum softening temperature was higher than 1,533 K. All the blended coals met the minimum requirements of pulverized coal ash melting point for blast furnace injection. The maximum temperature allowed by the experimental equipment was 1,673 K. When the temperature reached 1,673 K, there was no melting characteristic, and the characteristic temperature was recorded as ">1,673 K."

XRD analysis of the ash content of bituminous coal, anthracite, and blended coal was carried out by Yifan Chai et al. [1]. The main phase composition in anthracite coal ash is mullite and anorthite. The main phase composition in bituminous coal ash is calcite. The main phase composition in the mixed coal ash is anorthite. It shows that after mixing anthracite and bituminous coal at high temperature, mullite in anthracite ash reacted with anorthite in bituminous coal to form anorthite. The melting point of a crystal was related to the binding energy of the crystal. For ionic crystals, the lower the binding energy of the crystal, the lower the melting point of the substance. For the three crystals of mullite, anorthite, and anorthite, mullite has the largest crystal binding energy, the second crystal, and the smallest crystal. Therefore, the melting points of the three are in the order of mullite > anorthite > anorthite. When anthracite and bituminous coal were mixed, the mineral content of higher crystal binding energy had increased in the blended coal. It increased the melting point of the blended ash. This conclusion was verified through experiments and density functional theory calculations [1].

Figure 1 is the trend diagram of the ash melting characteristics of the blended coal mixed with BLT bituminous coal in different proportions as the percentage of bituminous coal ash content changes. It can be seen from Figure 1 that when bituminous coal with high ash melting point was added, the ash melting characteristics of blended coal improved. With the decrease of bituminous coal ash content, the deformation temperature, softening temperature, hemisphere temperature, and flow temperature of blended coal increased. It shows that blended coal had obvious improvement effect on ash fusion characteristics of bituminous coal with low ash melting point.

Under different mixing ratios, the effects of three kinds of anthracite mixed with BLT bituminous coal on the ash melting characteristics of blended coal are shown in Figure 2. The ash melting characteristic temperature of ">1,673 K" was plotted at 1,673 K like in Figure 1. It can be seen from Figure 2 that in comparison of the ash melting point characteristic temperature (softening temperature ST) data of pulverized coal, the ash melting point (softening temperature ST) of the blended coal with YQ anthracite was higher than that of the blended coal samples with JZ and LA pulverized coal under the same blending ratio conditions in all four blending ratios. It shows that the dispensing of YQ anthracite had the most significant effect on improving the ash melting characteristics of the blended coal. Among them, when YQ anthracite was mixed with BLT bituminous coal according to the ratio of 5:5, 6:4, 7:3, and 8:2, the ash melting points of the blended coal were 1,601, 1,639, 1,652 K, and more than 1,673 K, compared with the ash melting points of BLT bituminous coal, respectively, the ash melting points of the blended coal were increased by 98, 136, 149, and more than 170 K, respectively. Therefore, it can be concluded that when bituminous coal was selected for blast furnace smelting, the ratio of bituminous coal and anthracite with high ash melting point was less than 50%, the ash melting point of the blended coal was above 1,573 K, which can meet the needs of blast furnace injection.

3.2 Prediction of ash melting characteristics

The ash melting characteristic temperature experimental measurement results and the ash complete melting temperature values of single coal and mixed coal calculated by FactSage thermodynamic calculation software are shown in Table 6. It can be seen from Table 6 that the calculated value of ash complete melting temperature was greater than

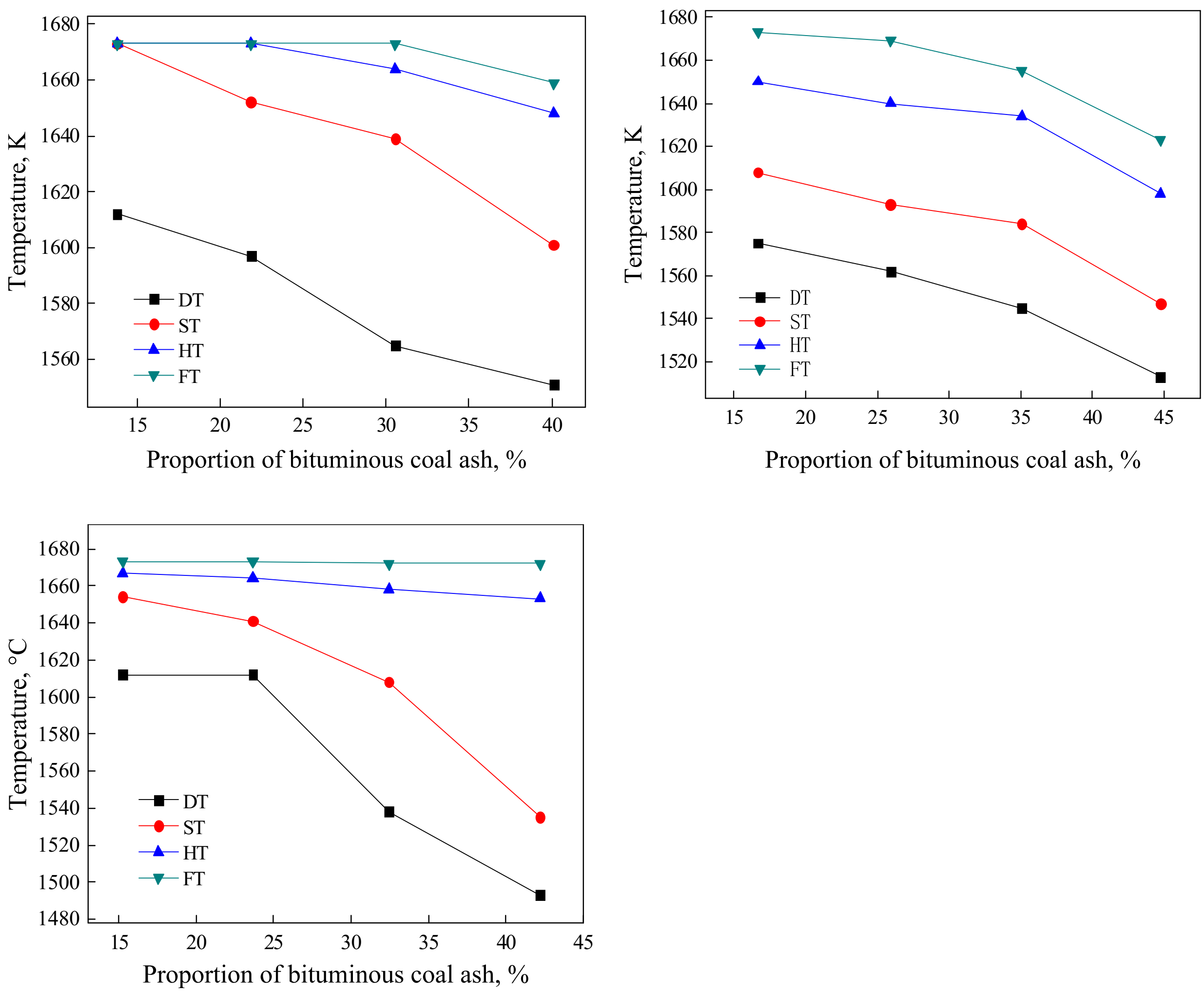

Figure 1: Relationship between ash partial melting characteristics of blended coal and ash content of high calcium bituminous coal: (a) YQ mixed with BLT, (b) JZ mixed with BLT, and (c) LA mixed with BLT (Note: The ash fusion characteristic temperature of ">1,673 K" is plotted with the value of "1,673 K.").

the experimental ash melting characteristic temperature. This is because the calculated value was the temperature when the solid components in ash were all converted into liquid phase, and the experimental process cannot ensure that the solid phase was completely converted into liquid phase. Because the limit temperature of the experimental equipment was not high enough, the ash melting state above 1,673 K cannot be judged by experiments. And in the process of ash melting characteristic experiment, the equipment needed to be heated and cooled, and the experiment period was long. This is also an advantage of phase equilibrium calculation compared to experimental determination, i.e., it can efficiently, quickly, and accurately calculate the temperature at which the melt completely melts at high temperatures.

Li's study [21] found that the ash liquid temperature can be used to predict ash melting temperature, and it fits a linear relationship ($T_M = 0.75T_C + 216.25$). T_M is the ash melting temperature and T_C is the ash complete melting temperature calculated by software. Shu used FactSage Software to study the effects of Ca and Si on the melting characteristics of coal ash [22]. Song used FactSage Software to study the effects of CaO, Fe_2O_3, and MgO on the melting characteristics of coal ash [23,24]. Liu used FactSage Software to calculate the coal ash melting temperature in a reducing atmosphere [25]. Van Dyk used FactSage Software to study the effects of Al, Si, and Ti on the melting characteristics of coal ash [26]. These scholars used the FactSage software method to calculate the liquid phase temperature of coal ash and predict the ash melting temperature. Select the softening temperature in the experimentally measured ash melting characteristic temperature and the calculated ash complete melting temperature to perform linear regression from Table 6. The

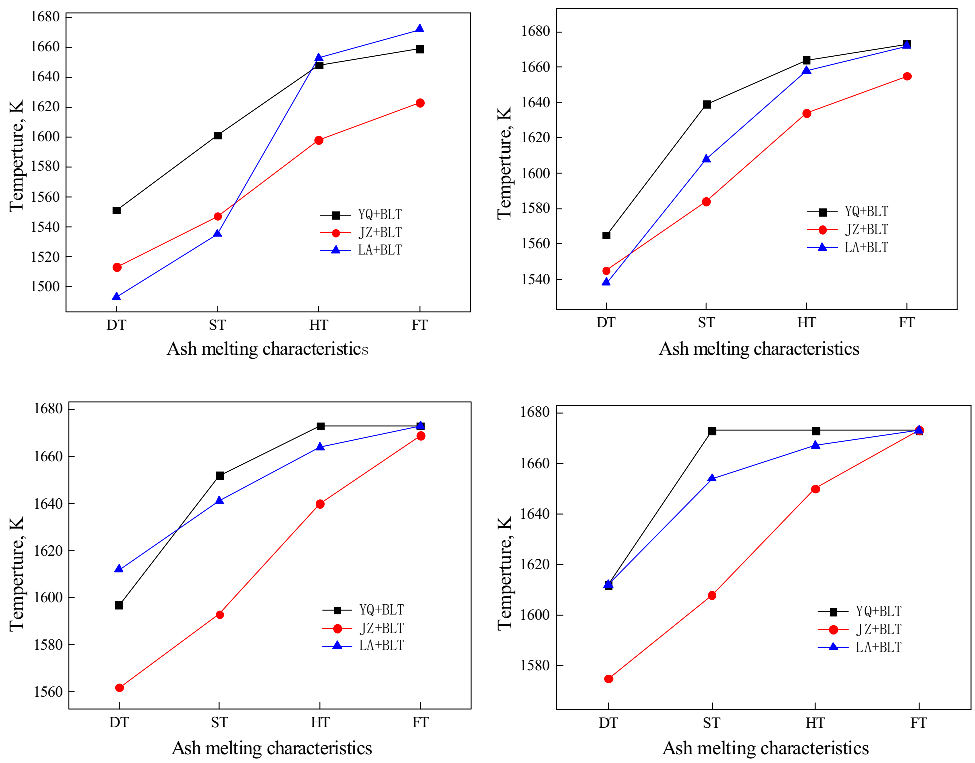

Figure 2: Variation of four kinds of ash melting characteristic temperature with coal type under various ratio conditions: (a) mixed ratio = 5:5, (b) mixed ratio = 6:4, (c) mixed ratio = 7:3, and (d) mixed ratio = 8:2.

Table 6: Calculation value of complete melting temperature of coal ash and experimental value of melting characteristic temperature

Code	Calculation of complete melting temperature (K)	Ash melting characteristic temperature (K)			
		DT	ST	HT	FT
BLT	1,695	1,465	1,503	1,525	1,538
YQ	2,007	>1,673	>1,673	>1,673	>1,673
JZ	1,961	>1,673	>1,673	>1,673	>1,673
LA	1,977	>1,673	>1,673	>1,673	>1,673
A1	1,749	1,551	1,601	1,648	1,659
A2	1,762	1,565	1,639	1,664	>1,673
A3	1,847	1,597	1,652	>1,673	>1,673
A4	1,918	1,612	>1,673	>1,673	>1,673
B1	1,746	1,513	1,547	1,598	1,623
B2	1,747	1,454	1,584	1,634	1,655
B3	1,758	1,562	1,593	1,640	1,669
B4	1,841	1,575	1,608	1,650	>1,673
C1	1,751	1,493	1,535	1,653	1,672
C2	1,744	1,538	1,608	1,658	1,672
C3	1,792	1,612	1,641	1,664	>1,673
C4	1,867	1,612	1,654	1,667	>1,673

relationship between the obtained coal fly ash melting point and the ash complete melting temperature calculated by the software is shown in Figure 3.

In the ash fusion characteristic experiment, the characteristic temperature above 1,673 K cannot be measured because of the equipment. Therefore, when performing linear regression, exclude the four groups of data with ash melting point experimental values greater than 1,673 K, “YQ,” “JZ,” “LA,” and “A4.” Through linear fitting, the new relationship between the melting point of coal ash

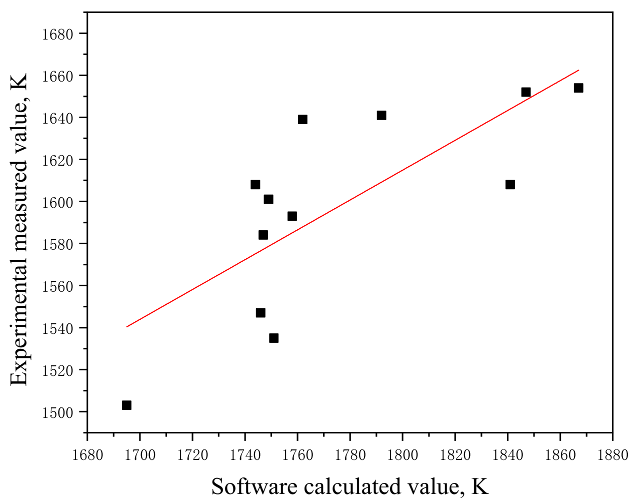

Figure 3: Relationship between software calculation of ash complete melting temperature and experimental measurement of ash melting point.

and the calculated value of the ash liquid phase temperature is obtained as: $T_{ST} = 0.7098T_C + 257.98$. Here T_{ST} is the ash melting point of pulverized coal and T_C is the complete melting temperature of pulverized coal ash calculated by the software. By comparison, the calculation formula was similar to the literature, indicating that there is a certain relationship between the ash melting point of pulverized coal and the complete ash melting temperature of pulverized coal. The ash melting point can be predicted efficiently and conveniently by FactSage software. It avoids cumbersome ash melting characteristic experiments and does not consider the limit temperature of the experimental equipment, which provides a new method for the prediction of coal ash melting temperature.

4 Conclusion

(1) When anthracite with high ash melting point was added, the ash melting characteristics of the blended coal were improved. As the proportion of bituminous coal ash decreased, the deformation temperature, softening temperature, hemispheric temperature, and flow temperature of the blended coal were all improved. Among them, the blending of YQ anthracite had the most significant effect on improving the ash melting characteristics of the blended coal. After adding different proportions of YQ anthracite, compared with the ash melting point of BLT bituminous coal, the ash melting point of the blended coal was increased by 98, 136, 149, and 170 K, respectively.

(2) The complete melting temperature of coal ash calculated by the FactSage thermodynamic calculation software had a certain relationship with the experimentally measured coal ash melting characteristics. The calculation of FactSage software can efficiently and conveniently predict the melting point of coal ash, avoiding tedious ash melting characteristic experiments and without considering the limit temperature of the experimental equipment, providing a new method for the prediction of coal ash melting temperature.

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Appendix

(1) Select the Equilib module (shown in Figure A1)

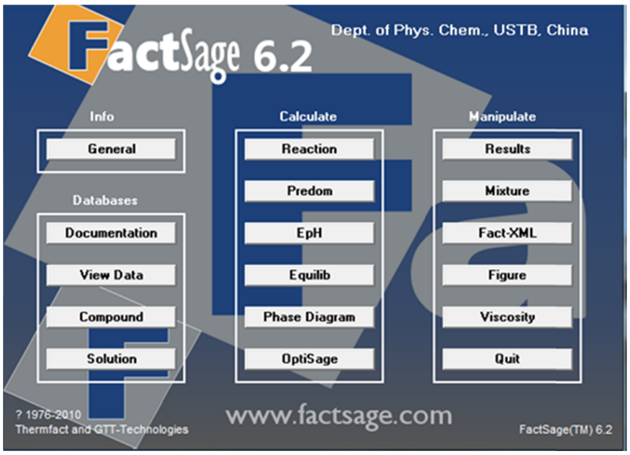

Figure A1: Module selection for calculating melting temperature.

(2) Select the FTOxid oxide database (shown in Figure A2)

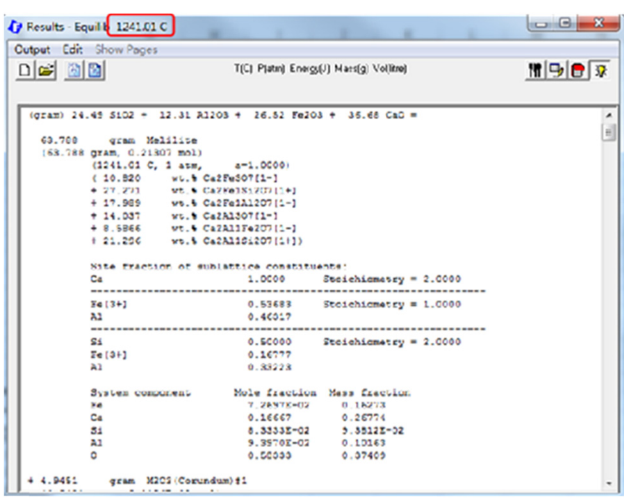

Figure A2: Database selection for calculating melting temperature.

(3) Input the composition of pulverized coal ash

The main oxides in the ash of selected pulverized coal samples were composed of SiO_2, Al_2O_3, Fe_2O_3, and CaO. After dividing the content of their components into hundreds of masses (shown in Table 4), input them in the interface of Figure A3 one by one.

(4) Select potential reaction products

The potential reaction products were selected at the calculation parameter setting interface. First, select all the solid pure substances in the product column. Second, click the "Select" key to select all solution phases retrieved from the FTOxid database.

That means select "Add all solutions from database" in the dialog box that pops up after clicking the "Select" key, and then click the "FToxid" key. This calculation step is shown in Figure A4. When calculating the "complete melting temperature," the mouse needs to be right-clicked to set the "FToxid-SLAGA" in the liquid phase "Solution species" to the "P" option (Precipitate target phase).

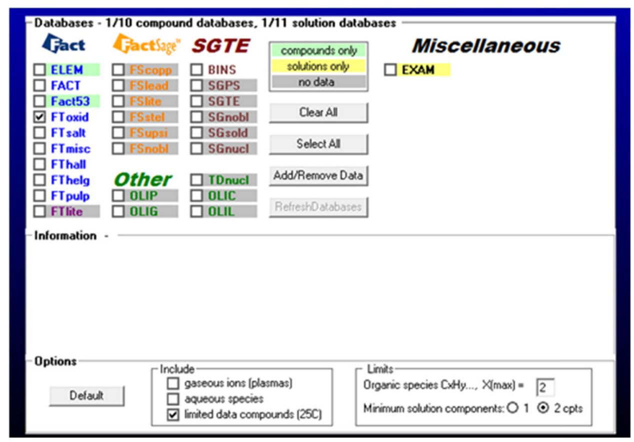

Figure A3: Calculation of the input of ash component in melting temperature.

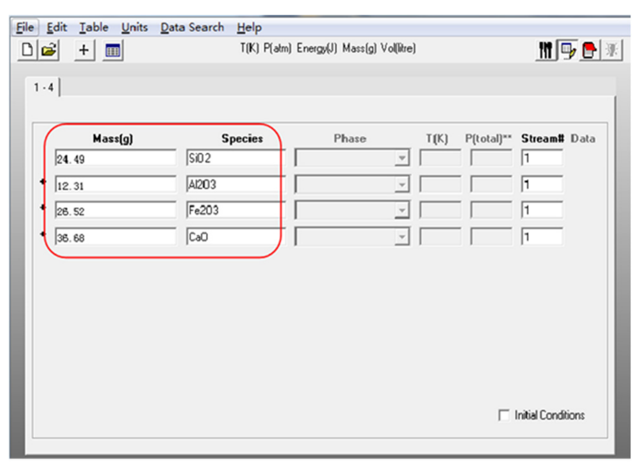

Figure A4: Selection of reaction products in calculation of melting temperature.

(5) Begin calculations and results

As shown in Figure A4, the "FToxid-SLAGA" in the liquid phase "Solution species" was set to the "P" option (Precipitate target phase). Then leave the temperature field in "Final Conditions" empty. The "complete melting temperature" of pulverized coal ash can be calculated by clicking "Calculation" after filling the pressure of the reaction system. The temperature in the red box in Figure A5 is the calculation result of "complete melting temperature."

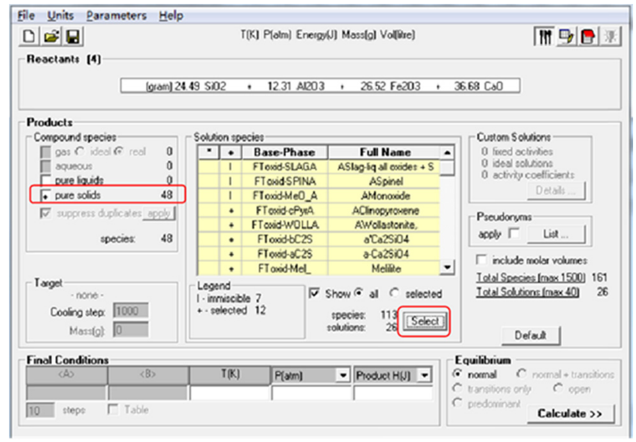

Figure A5: Calculation result of melting temperature.