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Experimental Study on the Effects of Chemical and Mineral Components on the Attrition Characteristics of Coal Ashes for Fluidized Bed Boilers

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ABSTRACT: In this paper, the attrition characteristics of ashes of 25 different coals used in circulating fluidized bed boilers were experimentally studied. The attrition characteristics were described by the attrition rate constant K_{af}, which was obtained by the static combustion and cold sieving method. The effects of 10 chemical components and 6 mineral components on K_{af} were evaluated by the gray relational analysis method. The main phases of the mineral components in the coal ashes were determined by X-ray diffraction, and anhydrite ($CaSO_4$), lime (CaO), quartz (SiO_2), hematite (Fe_2O_3), metakaolinite ($Al_2Si_2O_7$), and metaillite ($KAl_2AlSi_3O_{11}$) were identified. The results showed that, for a given mass fraction, the significance of the influence of the ash chemical components on K_{af} follows the sequence: CaO > MgO > SO_3 > P_2O_5 > Na_2O > Fe_2O_3 > K_2O > TiO_2 > Al_2O_3 > SiO_2. The significance of the influence of the ash mineral components on K_{af} follows the sequence: lime (CaO) > anhydrite ($CaSO_4$) > hematite (Fe_2O_3) > metaillite ($KAl_2AlSi_3O_{11}$) > metakaolinite ($Al_2Si_2O_7$) > quartz (SiO_2). In addition, the effect of the hardness of individual mineral components on K_{af} was also discussed.

1. INTRODUCTION

In the combustor of a coal-fired fluidized bed boiler, either a bubbling fluidized bed (BFB) or a circulating fluidized bed (CFB) one, there is a large amount of bed inventory. The quantity and particle size distribution (PSD) of the bed inventory play an important role in hydrodynamics, heat transfer, and combustion of the boiler, especially for the CFB one.[1] Both laboratory studies and practical experience showed that more than 90% of the bed inventory is made of the coal ashes. These particles can reside in the combustor for a long time, even up to 3−4 h. During this period, the ash particles rub against other particles, the walls, and internals of the combustor, resulting in detachment of fine pieces from the ash particles. Such a process is called ash attrition. Obviously, ash attrition characteristics is a key factor for the PSD of the bed inventory and thus the material balance and heat transfer in the combustor.[2]

Attrition characteristics of solid particles is often described by the attrition rate R_s or attrition rate constant K_{af}.[3,4] Mathematically, R_s is defined as

$$R_s = -\frac{1}{m}\frac{dm}{dt} \tag{1}$$

where m is the mass of solid particles within a given size range at a given time and dm/dt is the temporal mass variation rate. In a fluidized bed, K_{af} is expressed as

$$K_{af} = \frac{R_s}{u_g - u_{mf}} \tag{2}$$

where u_g is the superficial fluidizing velocity, m/s, and u_{mf} is the minimum fluidization velocity of the sample, m/s. ($u_g - u_{mf}$) is also called the excess fluidizing velocity, m/s.

On the basis of the above definitions, ash attrition characteristics are determined by the operational parameters of the boiler and the ash particle properties. Among the operational parameters, u_g is considered to be the most important one for a given ash because R_s is in direct proportion to $u_g - u_{mf}$[5] or the cube of u_g.[6] Among the particle properties, the size of the particles is of particular importance. Donsi et al.[7] found that the carbon attrition rate in their bubbling bed combustor is in inverse proportion to the average particle diameter. Wang et al.[8] found that K_{af} of the ashes from the same coal decays exponentially with increasing particle size.

Other than the physical properties, the chemical properties of the particles were also found to be important as well. Some researchers even attributed the difference of ash attrition characteristics to the different chemical components or the minerals in the ash. On the basis of the experiments of three coal ashes in a laboratory-scale CFB combustor, Winter and Liu[9] found that elements Si and Ca play an important role in ash attrition and the higher the Si or the lower the Ca content is, the less the ash is prone to attrition. Using the gray relational analysis (GRA) method to study the influence of mineral components on K_{af} of four coals, Wang et al.[8] found that the strength of the influence on K_{af} is in the following sequence: lime (CaO) > hematite (Fe_2O_3) > metakaolinite ($Al_2Si_2O_7$) > quartz (SiO_2). They also suggested that K_{af} should be related to the mineral components' hardness; however, no detailed correlation was given.

Though the existing studies showed the importance of chemical and mineral components on the ash attrition, experimental data are still very limited. In addition, so far, the

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Table 1. Proximate and Ultimate Analyses and LHV of Raw Coal Samples

sample	C_{daf} (%)	H_{daf} (%)	O_{daf} (%)	N_{daf} (%)	S_{daf} (%)	M_{ar} (%)	A_{ar} (%)	V_{ar} (%)	LHV_{ar} ($MJ \cdot kg^{-1}$)
BDG	53.75	6.47	38.26	0.96	0.56	2.49	66.83	17.50	4.15
BDS	77.89	4.92	14.83	1.46	0.90	26.91	24.09	18.97	19.30
DTE	75.17	5.15	18.08	1.00	0.59	10.33	51.98	16.43	11.03
DTH	50.37	2.94	5.94	0.67	0.74	1.09	40.03	14.29	19.04
DTR	80.76	4.94	12.57	1.36	0.37	8.92	11.42	30.86	24.92
Gardanne	51.39	3.17	10.19	1.14	3.30	5.78	25.03	30.33	18.49
G2900	75.07	4.98	18.32	1.17	0.46	11.53	43.4	19.56	12.51
G4100	80.13	4.97	13.21	1.26	0.43	13.56	16.81	27.20	21.41
HLBE	46.87	3.13	13.6	0.71	0.27	21.75	13.67	46.78	17.85
JCM	61.83	2.92	4.46	0.94	0.58	3.08	26.19	9.37	25.19
LA[a]	62.13	2.67	7.33	1.18	0.15	2.12	26.54	10.31	25.05
LY[a]	52.30	1.04	0.83	0.71	1.12	4.72	62.96	4.30	19.07
NJ	75.25	6.99	14.85	1.88	2.65	7.50	42.00	9.12	15.34
Peru	54.63	1.1	5.75	0.51	0.8	4.74	32.47	9.87	19.33
SH	57.98	3.32	18.62	0.44	0.72	3.68	17.29	31.42	21.73
SJZ	86.39	4.42	9.40	1.29	0.85	6.80	29.88	6.05	23.99
SLQB	80.78	4.01	10.93	1.21	3.06	0.77	32.36	11.87	21.68
SLQM	72.21	4.18	18.63	0.86	4.12	20.97	21.24	22.46	16.18
SWCS	78.71	4.14	15.79	0.87	0.49	16.51	7.86	24.96	23.27
SWFS	79.79	4.07	14.45	0.94	0.76	31.3	9.83	18.6	24.37
WD	80.68	5.28	11.51	1.42	1.12	9.95	20.35	28.04	21.96
WH	80.74	5.20	11.58	1.25	1.26	6.78	33.04	15.11	18.12
XLT[a]	36.72	1.87	12.59	1.01	1.66	9.88	4.95	49.07	12.43
YL	82.27	5.14	9.90	0.99	1.70	22.28	19.84	46.89	17.46
ZB	83.07	4.64	1.67	9.36	0.78	4.44	33.14	10.44	21.06

[a]Proximate and ultimate analyses are on an air-dried basis.

studies only focused on the effects of minerals made of Ca, Fe, Al, and Si, and nothing on the effects of minerals that made up other rich elements in the coal ash, such as Mg, Na, K, and S. Obviously, more comprehensive studies on the effect of chemical or mineral components on the ash attrition are needed.

In this paper, the attrition characteristics of a number of ashes from different coals are studied. The influences of 10 chemical components, including CaO, Fe_2O_3, Al_2O_3, SiO_2, MgO, Na_2O, K_2O, SO_3, TiO_2, and P_2O_5, and 6 minerals containing Ca, Fe, Al, Si, and S on the ash attrition are assessed. The effect of the mineral components' hardness on ash attrition characteristics is also discussed.

2. EXPERIMENTAL SECTION AND METHODOLOGY

2.1. Raw Coal Properties. In the present study, the coal ashes are made from 25 raw coals, 23 from China, 1 from France (Gardanne), and 1 from Peru. The coals are used in CFB boilers in different power plants. Their proximate and ultimate analyses as well as the lower heating values (LHVs) are given in Table 1. The coals include gangue, anthracite, lignite, bituminous, slime, and middling, nearly every type of coal used in the power generation industry.

2.2. Determination of Attrition Rate Constant. The attrition rate constant K_{af} was experimentally determined by the static combustion and cold sieving (SCCS) method that has been used in the previous studies.[2,10−12] In the SCCS experiment, a certain amount of raw coal was sieved into six narrowly sized groups. Samples of each group were incinerated individually in a muffle furnace at a temperature of 850 °C, close to the normal operational temperature of a CFB boiler. After the burnt-out time, which was preset upon the coal type and the size of the samples, the coal sample was burnt into coal ashes and collected. When it was cooled down, the collected ashes were then sieved by a shaker under a preset shaking amplitude of 2 mm. The sieving was stopped every 10 min. During each break, part of the ash was sampled out to measure the size distribution. At the same time, the cumulative undersize distribution of the particles was obtained. After a certain time, normally 30−60 min, the cumulative undersize distribution of all six groups became nearly identical, and the attrition process became steady.[2]

On the basis of the measured ash size evolution data in the above experiments, the sieving ash attrition rate constant K_{as} can be calculated by eq 3[11]

$$K_{as} = R_{as}/A^2 \tag{3}$$

where R_{as} is actually equal to R_s in eq 1. A is the sieving amplitude of the shaker, m.

Previous experimental study found that the solid attrition mechanism is similar in the fluidized bed and shaking system. By properly correlating the superficial velocity in the fluidized bed with shaking amplitude A in the shaker with respect to the system energy, K_{as} obtained by the SCCS method can be converted into K_{af} with acceptable accuracy.[2,11]

2.3. Evaluation Method for Chemical/Mineral Component Effect. To evaluate the effect of chemical or mineral components on K_{af}, the gray relational analysis (GRA) was adopted in the present study. The GRA is part of gray system theory that has been developed for about three decades.[13] It can be used to effectively solve the complicated interrelationships between multiple factors and variables through the optimization of gray relational grades (GRD).[14] The detailed calculation in the GRA method was described in a number of papers.[14−16]

In the present study, the mass fraction of the same chemical or mineral component of all ashes was set as a compared sequence and K_{af} was set as the reference sequence. Therefore, the size of each sequence was 25 × 1 and the number of compared sequences was the number of chemical or mineral components. Experimental data in every sequence were first self-normalized in the range between zero and one, and then GRD γ_i, which described the degree of dependency of the ith chemical or mineral component on K_{af}, was calculated from the normalized data to correlate the reference sequence (K_{af}) and the compared sequence (a chemical or mineral component).

If $\gamma_k > \gamma_1 > ...\gamma_p$, then the kth chemical or mineral component had the most significant influence on K_{af}, the first component followed the kth one, and the pth component had the weakest influence.

3. RESULTS AND DISCUSSION

3.1. Attrition Constants of the Coal Ashes. Figure 1 shows the typical variation of the cumulative mass fraction of the undersize particle sizes in various sieving times with the raw coal size of 1000−2000 μm for the WD ash. The difference of the distribution curves becomes negligible after 50 min. According to the SCCS method, K_{af} of each coal ash with a raw coal size of 1000−2000 μm was determined after the curves collapsed.

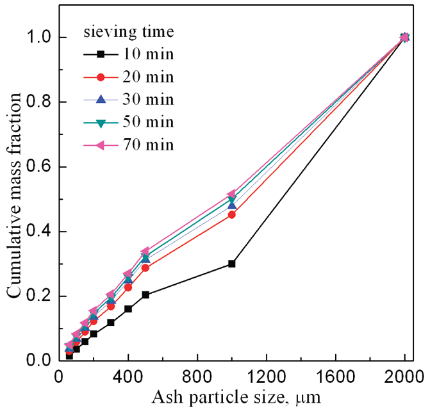

Figure 1. Typical cumulative undersize distribution of ash particle size (WD coal ash, raw size = 1000−2000 μm).

The chemical composition and the K_{af}'s determined by the SCCS method of coal ashes values are listed in Table 2.

As shown in Table 2, the values of K_{af}'s vary from 1.15 × 10^{-6} m^{-1} (for DTE ash) to 2.0 × 10^{-5} m^{-1} (for Gardanne ash). The ashes having the largest K_{af}'s are Gardanne, XLT, and HLBE ash. They are also the three coal ashes having the largest mass fraction of CaO and the smallest mass fractions of SiO_2 and Al_2O_3.

However, the relationship between chemical components and the K_{af} of other ashes is ambiguous, as shown in Figure 2.

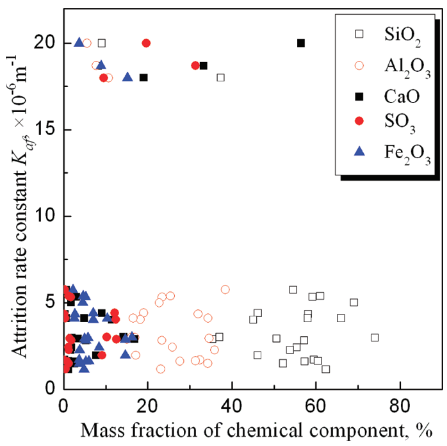

Figure 2. Relation between the mass fraction of the main chemical components and the attrition rate constant.

This might be due to the cross-interaction of multiple components. It makes the determination of the relationship

Table 2. Chemical Analysis (wt %) and Attrition Rate Constant for Tested Coal Ashes

sample	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	K_2O	Na_2O	TiO_2	P_2O_5	K_{af} ($\times 10^{-6}$ m^{-1})
BDG	54.54	38.46	2.18	0.54	0.29	0.27	0.32	0.09	2.45	0.48	5.76
BDS	50.58	34.57	4.94	3.44	0.54	1.44	1.20	0.12	2.12	0.37	2.93
DTE	62.36	23.13	4.85	1.14	1.70	0.43	3.90	1.01	1.04	0.10	1.15
DTH	59.49	32.38	3.64	0.16	0.25	0.05	1.18	0.14	2.33	0.11	1.70
DTR	57.17	23.36	5.23	5.27	1.36	1.85	3.27	0.54	1.03	0.34	2.82
Gardanne	9.04	5.59	3.65	56.49	1.35	19.68	0.27	0.10	0.23	0.60	20.0
G2900	57.26	27.65	5.24	2.90	1.64	0.76	2.40	0.52	1.08	0.20	1.61
G4100	59.14	23.39	5.21	2.96	1.78	1.73	3.73	0.45	1.06	0.06	5.33
HLBE	37.36	10.66	15.16	19.02	4.75	9.52	0.53	1.12	0.95	0.06	18.0
JCM	57.98	34.32	2.56	0.54	0.29	0.38	0.90	0.31	2.13	0.17	4.10
LA	52.17	34.30	3.65	2.54	0.64	1.23	2.33	0.62	1.70	0.33	1.49
LY	55.44	25.85	8.41	1.78	1.87	1.11	3.31	0.28	1.23	0.21	2.42
NJ[a]	74.00	16.93	5.80	1.95	0.58		0.44	0.04	0.16	0.10	2.98
Peru	58.16	31.67	2.68	0.25	0.66	0.14	3.42	0.41	1.86	0.14	4.34
SH	46.16	19.04	7.01	9.04	3.20	12.11	1.71	0.35	0.80	0.06	4.40
SJZ[a]	60.46	31.38	6.02	1.05	0.45		0.44	0.03	0.06	0.10	1.63
SLQB	53.81	35.90	3.77	1.61	0.17	1.22	0.77	0	2.48	0.04	2.26
SLQM	46.08	17.14	14.64	7.69	2.19	9.10	1.55	0.47	0.65	0.06	1.96
SWCS	35.39	15.05	14.65	16.84	1.28	12.58	0.81	1.80	0.64	0.03	2.89
SWFS	37.01	16.19	16.25	14.21	1.80	10.27	1.11	1.03	0.62	0.07	3.01
WD	60.99	25.41	4.60	1.69	1.00	1.16	2.83	0.50	1.21	0.20	5.39
WH[a]	65.91	16.50	10.35	4.89	1.23		0.53	0.14	0.23	0.21	4.10
XLT	8.20	7.74	8.86	33.25	9.04	31.36	0.39	0	0.42	0.12	18.7
YL	45.06	18.25	7.03	11.49	2.14	12.38	1.58	0.54	0.85	0.04	4.02
ZB[a]	69.05	22.73	4.53	1.71	1.08		0.69	0.04	0.08	0.09	5.00

[a]Ashes (four types) were chemically analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES). SO_3 was not measured, and the content of SiO_2 was obtained from the following equation: $SiO_2 = 100 - Al_2O_3 - Fe_2O_3 - CaO - MgO - K_2O - Na_2O - TiO_2$ wt %. The chemical analyses of the other ashes were carried out by X-ray fluorescence (XRF).

Table 3. GRDs of Ash Chemical Components for Their Attrition Rate Constant

CaO	MgO	SO_3	P_2O_5	Na_2O	Fe_2O_3	K_2O	TiO_2	Al_2O_3	SiO_2
0.84	0.82	0.79	0.75	0.74	0.71	0.63	0.60	0.51	0.45

between a single component and K_{af} difficult. Nevertheless, it can still be seen that the high concentrations of SiO_2 and Al_2O_3 generally correspond to a low K_{af}, causing the ash to be more attrition resistant. The high CaO concentration is most likely to have a high K_{af}. The Fe_2O_3 is not necessary to be a direct index for K_{af}. In addition, it can be seen that, when SO_3 is high, the K_{af} intends to be high.

3.2. Influence of Chemical Components on Ash Attrition. The GRDs of chemical components for K_{af} obtained from the above GRA method are listed in Table 3. The significance of the influence of ash chemical components has the sequence: CaO > MgO > SO_3 > P_2O_5 > Na_2O > Fe_2O_3 > K_2O > TiO_2 > Al_2O_3 > SiO_2. This sequence is in accordance with the phenomena presented in the last section. Namely, the ash with the largest amount of CaO and the smallest amount of SiO_2 and Al_2O_3 has the largest value of K_{af}. It also agrees with Winter and Liu's[9] results that the ash with the higher Si content is less prone to attrition and the ash with the higher Ca content is more prone to attrition.

3.3. Main Mineral Phases in Coal Ashes. Although the composition of ash is usually expressed as oxides shown in Table 2, the mineral form is the practical existence form of the chemical component in ash. Thus, it is necessary to assess the influence of mineral phases on the ash attrition characteristics. There are more than 20 types of mineral phases in raw coals, and they can be generally categorized into five groups, namely, silicates (quartz, kaolinite, illite, plagioclase, etc.), oxides and hydroxides (hematite, goethite, etc.), sulphates (gypsum, anhydrite, etc.), carbonates (calcite, dolomite, siderite, etc.), and sulphides (pyrite, marcasite, etc.).[17,18] After the coal is burnt in the boilers, some new phases could be formed and the total number of mineral phases in ashes could be over 50.[19] Therefore, it is important to identify which are the main mineral phases, such that the influence of minerals on ash attrition characteristics can be effectively and feasibly evaluated.

In the present study, four typical ashes generated from XLT lignite, DTH midding, SH bituminous, and LY anthracite were selected to identify the main mineral phases in the ash. Figure 3 depicts the results of X-ray diffraction (XRD) analyses on mineral phases for these ashes.

For XLT ash, in which there are large amounts of CaO and SO_3, anhydrite ($CaSO_4$) is the predominant phase. Anhydrite is also identified in SH ash, in which the mass fraction of CaO and SO_3 is about 10%, and in LY ash, in which the mass fraction of CaO and SO_3 is about 1%. No anhydrite is found in DTH ash due to the low mass fractions of CaO and SO_3 (<0.2%). As shown in Figure 3a,d, the rest of the CaO other than that in anhydrite can exist in the form of lime (CaO) in XLT and LY ashes. In SH ash, the mass ratio of SO_3 to CaO is about 1.34, approaching the equivalent ratio of 1.43 in anhydrite, so there is no lime detected.

As shown in Figure 3, hematite and quartz are the major phases detected by XRD in all ashes listed in Table 2. These ashes have a relatively high mass fraction of Fe_2O_3 and SiO_2. Anhydrite and lime can be significant but depend on the mass fraction of CaO and SO_3. Corundum (Al_2O_3) is found in SH ash. Actually, silicates should be the major phases for the element Si and the predominant phase for the element Al.[17,18]

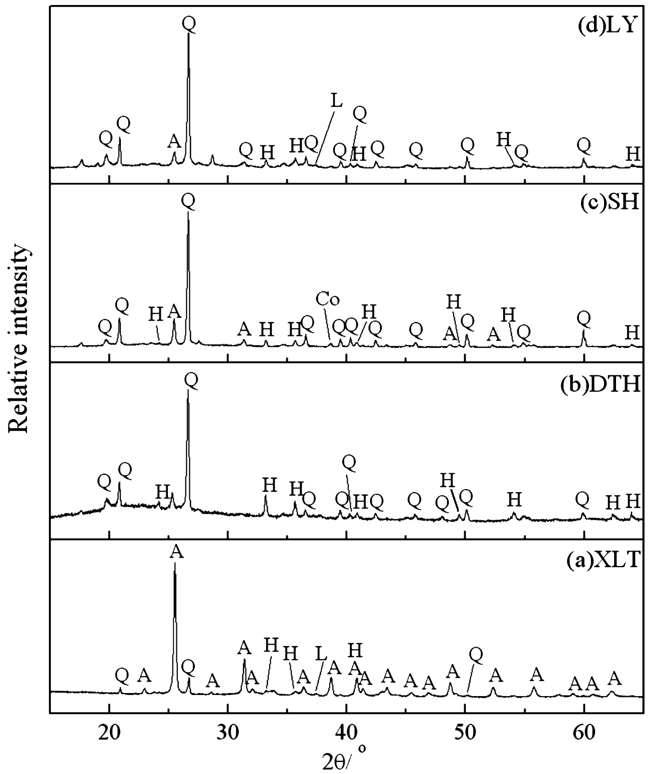

Figure 3. XRD patterns of four coal ashes generated at 850 °C. Mineral and phase abbreviations: A, anhydrite ($CaSO_4$); Co, corundum (Al_2O_3); H, hematite (Fe_2O_3); L, lime (CaO); Q, quartz (SiO_2).

The difficulty for silicate identification by XRD in this study might be caused by the complicated characteristic peaks and poor crystallinity of silicates. Metakaolinite ($Al_2Si_2O_7$) and metaillite ($KAl_2AlSi_3O_{11}$) could also be the major phases in ashes generated at the temperature of 850 °C.[17,18]

On the basis of the above analyses and discussion, anhydrite ($CaSO_4$), lime (CaO), quartz (SiO_2), hematite (Fe_2O_3), metakaolinite ($Al_2Si_2O_7$), and metaillite ($KAl_2AlSi_3O_{11}$) were identified as the main phases in coal ash.

3.4. Influence of Mineral Components on Ash Attrition Characteristics. The GRDs of the six mineral components for K_{af} obtained from the GRA method are listed in Table 4. The relative significance of the influence of the minerals on K_{af} is lime (CaO) > anhydrite ($CaSO_4$) > hematite (Fe_2O_3) > metaillite ($KAl_2AlSi_3O_{11}$) > metakaolinite ($Al_2Si_2O_7$) > quartz (SiO_2).

Table 4. GRDs of Ash Mineral Components for Their Attrition Rate Constant

lime CaO	anhydrite $CaSO_4$	hematite Fe_2O_3	metakaolinite $Al_2Si_2O_7$	metaillite $Al_2AlSi_3O_{11}$	quartz SiO_2
0.82	0.79	0.71	0.60	0.58	0.53

The influence of minerals on K_{af} can probably explained by their hardness or attrition resistance listed in Table 5.

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Table 5. Mineral Hardness (on the Mohs Scale)[20]

quartz SiO_2	hematite Fe_2O_3	anhydrite $CaSO_4$	lime CaO	kaolinite $(Al_2Si_2O_5(OH)_4)$	illite $KAl_4[Si_7AlO_{20}](OH)_4$
7.0	6.0	3.5	1.8	2.3	1.5

Because it is difficult to find the hardness values of metakaolinite ($Al_2Si_2O_7$) and metaillite ($KAl_2AlSi_3O_{11}$), the values of kaolinite ($Al_2Si_2O_5(OH)_4$) and illite ($KAl_4[Si_7AlO_{20}](OH)_4$) are used instead, respectively. According to Tables 4 and 5, the increase of hardness will reduce the GRD values except for the minerals metakaolinite (kaolinite) and metaillite (illite). Moreover, the increase in the mass fraction of the softer minerals will significantly increase K_{af}, namely, more prone to attrition.

The discrepancy of metakaolinite (kaolinite) and metaillite (illite) could be due to the improper mineral phase selection for elements Al and K. The complicated transformation process for various raw mineral phases makes the determination of the final minerals in ash difficult. It was found that corundum (Al_2O_3) is formed in the present study (Figure 3c) and the literature for some coals.[17,18] Corundum is a much harder mineral compared with others, with a Mohs hardness of 9.0.[20] Thus, it can increase the ash attrition resistance and decrease the value of K_{af}.

4. CONCLUSIONS

On the basis of the experimental study on attrition characteristics of ashes of 25 different coals used in circulating fluidized bed boilers by the SCCS method, one can conclude that high SiO_2 and Al_2O_3 contents are prone to have low ash attrition rate constants K_{af}'s, and thus high attrition resistance. The assessment of the influences of 10 chemical components on K_{af} by the GRA method revealed that, with the same mass fraction, the significance of the influence of the ash chemical components on K_{af} follows the sequence: CaO > MgO > SO_3 > P_2O_5 > Na_2O > Fe_2O_3 > K_2O > TiO_2 > Al_2O_3 > SiO_2. XRD analyses on mineral phases showed that the chemical components exist in different forms in the coal ash, and the main phases include anhydrite ($CaSO_4$), lime (CaO), quartz (SiO_2), hematite (Fe_2O_3), metakaolinite ($Al_2Si_2O_7$), and metaillite ($KAl_2AlSi_3O_{11}$). For these six mineral components, GRA analyses showed that the significance of the influence on K_{af} follows the sequence: lime (CaO) > anhydrite ($CaSO_4$) > hematite (Fe_2O_3) > metaillite ($KAl_2AlSi_3O_{11}$) > metakaolinite ($Al_2Si_2O_7$) > quartz (SiO_2). Further discussion confirmed that the hardness of an individual mineral component and its mass fraction in the coal ash could be the dominant control factor for the ash attrition characteristics.

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