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Mechanistic studies on the slagging propensity in low-rank coal combustion



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ABSTRACT

Utilizing low-rank coals in power plants takes a risk of slagging, especially when burning coals rich in alkali and alkaline earth metallic (AAEM) elements. To predict the slagging propensity of low-rank coals, this work develops a novel approach for accurately measuring the sticking probability of thermally stable slag samples based on the well-controlled flat-flame system. We collect deposit samples (PSD) severely accumulated on the platen superheater surface of a 660 MW boiler co-firing two low ranks (alkali-rich Phi-coal: iron-rich Ind-coal = 15.2 wt%: 84.8 wt%). It is found that the existing theoretical models can hardly reproduce the measured sticking probability at elevated temperatures. We thus propose an Sshaped nonlinear correlation of ash sticking probability with the melt fraction using two tuning parameters. The fully-sintered PSD slag is mainly composed of refractory anorthite (CaAl₂Si₂O₈) and hematite (Fe₂O₃), which are largely determined by the dominant Ind-coal ash fed into the boiler. However, the excessive accumulation of PSD can only be attributed to the much stickier Phi-coal ash rich in lowmelting-point, dispersed Na-aluminosilicates. Combining the measured sticking probability and physicochemical properties of the slag and ash samples, we reveal a positive correlation of the ash sticking probability with the sodium content of the samples, almost regardless of the contents of alkaline earth metals and iron. It highlights the leading role alkali metals play in slagging. In contrast, iron exists as discrete hematite (Fe_2O_3) in the slag and is less effective than AAEM in forming slags on the platen superheater. Our work provides a more reliable method and criterion for evaluating the slagging propensity of fully/partially-molten particles in solid fuel combustion.

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1. Introduction

Slagging of molten non-combustible matters during solid fuel combustion is ubiquitous in a variety of applications, including the coal/biomass-fired power plant [1] and the rocket motor fueled by solid propellants [2, 3], leading always to the performance degradation and damage to surfaces. In the coal-fired boiler operation, there are many other adverse effects of slagging. For instance, slagging elevates the flue gas exit temperature from the economizer and causes problems for running selective catalytic reduction (SCR) systems downstream. Modern utility boilers, equipped with tens to hundreds of soot blowers, can effectively clean the loosely-fouled convective heating surfaces [4]. However, the more densely-deposited slag on the radiant surface is less vulnerable to the attack from the compressed steam. Consequently, severe slagging accumulated around the burners 'eyebrows', at the upper fur-

* Corresponding author. E-mail address: lishuiging@tsinghua.edu.cn (S. Li). nace arch, and on the platen superheater surface are frequently encountered in practical boilers [5, 6].

Two emerging trends tend to aggravate slagging in power plants. First, the demand for highly flexible operation of coal-fired units results in weakened flame stability and less flowrates at reduced outputs, both facilitating the deposition of molten coal/ash particles [7, 8]. Secondly, more and more power plants are being tempted to burn low-cost coals, often of low-rank (or even biomass), to fully or partly replace the designed coals [9-11]. Unfortunately, slagging stands out for many low-rank coals, including the Victorian brown coal in Australia [12], Zhundong coal in Northwestern China [13], and several coal samples used in Southeast Asia [14]. The higher contents of AAEM in these coals play a significant role in the initiation of slag layers by forming sticky, low-melting-point eutectics [15–17]. It highlights the difference between the low-rank coal and common bituminous coal, because for the latter iron is believed a major inducer for slagging [18, 19]. Considering the vast demand for low-rank coals in developing countries, it calls for elaborate efforts to demonstrate the AAEMassociated slagging mechanism, and to provide insights on the

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Table 1

Loai properties.		
Samples	Phi-coal	Ind-coal
Proximate analysis (wt%, dry basis)		
Volatile matter	38.45	50.64
Fixed carbon	37.70	44.57
Ash	23.84	4.79
Ash compositions (wt%)		
SiO ₂	53.09	32.32
Al ₂ O ₃	23.52	17.44
CaO	3.95	10.97
Na ₂ O	1.76	0.23
Fe ₂ O ₃	7.07	30.94
MgO	2.10	2.36
SO ₃	4.84	3.20
K ₂ O	2.06	0.92
TiO ₂	1.05	0.87
$R_{B/A}^{a}$	0.22	0.90

 a basic to acidic oxides ratio, $R_{B/A}{=}~(Na_2O{+}CaO{+}Fe_2O_3{+}MgO{+}K_2O)/(SiO_2{+}Al_2O_3{+}TiO_2).$

slagging propensity for a wide range of operating conditions and varying fuel sources [20].

To conveniently evaluate the slagging propensity of various fuels, some empirical indices based on the coal ash composition, such as the basic to acidic oxides ratio $(R_{B/A})$ [21], silica to alumina ratio $(R_{Si/Al})$ [22], and ash fusion temperature [23] have been traditionally used in the community. However, these methods are reportedly unreliable when forecasting ash deposition of different coal samples with an accuracy lower than 50% [24]. To tackle this issue, mechanistic investigations have delved into the dynamics of slagging, which contains the impaction and sticking/rebound of incoming particles. The impaction efficiency is determined by the flow Reynolds number and particle Stokes number (the ratio of the particle relaxation time to the fluid characteristic time) [25]. Nevertheless, the sticking probability of the impacting particle is a key factor yet to be fully understood [26]. Several phenomenological models have been proposed in the literature. A widely-used formula determines the sticking probability from the compositionrelated *particle viscosity* [27]. The model contains a free parameter termed the 'reference viscosity' which ranges across several orders of magnitude (8-10⁸ Pa•s) in Refs. [28, 29]. Another strategy is to assume the ash sticking probability to linearly increase with the particle melt fraction ranging from 10% to 70% [30]. Both types of models can be simplified to a '0/1-gate' with a threshold of viscosity or melt fraction [31, 32], and have been further extended by involving the incoming kinetic energy [32, 33]. However, these models often give inconsistent predictions [4, 34, 35], and more accurate and mechanistic experiments are needed to illuminate the sticking behaviors.

For this purpose, we note numerous studies on hightemperature slagging were performed in experimental facilities of various scales [5, 36-38]. Generally, slag characterization from the full-scale boiler provided information on the real problem of concern, especially for alkali-[39] and iron-rich [18, 40] coals. Based on this, studies on lab-scale setups achieved better controls of the ambience (temperature, atmosphere, etc.) [17, 36]. While kilowattscale furnaces are quite successful in simulating fouling conditions after coal burnout, it is still challenging to precisely adjust the temperature and residence time for the early-stage slagging. By contrast, the bench-scale Hencken flat-flame burner is able to flexibly adjust the flow velocity, ambience temperature, and atmosphere [20, 41]. Thus, it could well mimic the local environment of slagging in the practical boiler by entraining coal particles [42, 43] or even ash particles of similar sizes and compositions to the slag. Indeed, one may wish to generate entrained particles from real slag samples with chemically-stable compositions after being exposed

to the high temperature for a sufficiently long time. This approach, by incorporating the full-scale sampling and bench-scale investigation, is expected to reveal the slagging mechanism of low-rank coals and serve as a working criterion to examine sticking probability models. But it was rarely reported before.

In this work, we thoroughly characterized the deposit samples collected from different heat transfer surfaces of a 660 MW pulverized coal boiler. We investigated the phase transition and melting behaviors of the deposits under elevated temperatures by thermodynamic simulations. The effect of AAEM on slag formation is revealed. Furthermore, the high-temperature sticking probabilities of slag particles sampled from the platen superheater and the water wall were derived in a well-designed bench-scale flat-flame burner. The results indicate a nonlinear relation with the melt fraction of impacting particles. The findings help elucidate the contribution of the preferential distribution of mineral species (mainly AAEM) to low-rank coal slagging.

2. Methodology

2.1. Investigated unit and deposit samples

As sketched in Fig. 1a, we sampled different deposits from a 660 MW, supercritical coal-fired boiler in Southeastern China. The boiler had an unscheduled shutdown after SCR splitting due to excessively high flue gas temperatures. This was most likely caused by the severe slagging on the platen superheater surface after over 75 days' operation (see Fig. 1a). Comparatively, slagging/fouling on other heating surfaces was slighter. The deposits of four parts (see Fig. 1a), including the water wall (WW), the platen superheater (PS), the pendant final superheater (FS) and the pendant hightemperature reheater (HR), were sampled and analyzed. The bulky slag in the outer layer of those regions (not adjacent to the heating surface) was knocked and collected for its good representation of the deposit accumulated from inertial impaction of coarse ash particles. Hereafter, along the flowsheet of the boiler, the deposit samples from these parts are successively denoted as WWD, PSD, FSD and HRD. In the high-temperature zones (i.e., 1080 °C or higher), the dark, bulky slags WWD and PSD are fully-sintered and difficult to break. In contrast, the partially-agglomerated FSD is akin to compacted sand and can be easily destroyed by external forces. The presence of HRD is only in small quantities and could shed with the corrosion of tubes. Figure 1b presents the ranges of the flue gas temperature for the PS (1080–1310 °C), FS (950–1080 °C) and HR (850–950 °C) surfaces when the boiler operates at different loads.

2.2. Coal properties

The coal burnt in the boiler includes the mixture of Philippine coal (Phi-coal) and Indonesian coal (Ind-coal) imported from Southeast Asia. The average blending ratio Phi-coal: Ind-coal = 15.2wt%: 84.8 wt% during the last 19 days before shutdown. Pulverized raw coal samples were collected from the plant, with Table 1 presenting the coal properties. The size distribution of the raw coal samples was measured by a Mastersizer 2000 Particle Size Analyzer (Malvern Panalytical Inc.), as shown in Fig.2. In particular, the Phi-coal ash contains higher contents of Na and K (1.31 wt% and 1.71 wt%, respectively), which can be identified as high alkali coal, whereas the Fe content in Ind-coal is up to 21.66 wt%. For the mineralogical properties, Fig. 3 shows the X-ray diffraction results of the coal ashes prepared at 600 ± 5 °C. The Ind-coal ash is dominated by quartz and hematite, whereas the Phi-coal ash is mainly composed of quartz, calcium sulfate and nepheline. Notably, the basic to acidic oxides ratios $R_{B/A}$ of Phi-coal ash and Ind-coal ash



Fig. 1. (a) Schematic of the 660 MW wall-fired boiler and the morphology of sampled deposits; (b) flue gas temperatures of sampling positions under different loads.



Fig. 2. Volumetric particle size distribution of pulverized Phi-coal and Ind-coal.



Fig. 3. XRD patterns of the coal ash samples. Q: quartz (SiO₂), He: Hematite (Fe_2O_3), An: Anhydrite (CaSO₄), N: Nepheline (NaAlSiO₄)

are 0.22 and 0.90, respectively. This preliminary estimation indicates a much higher slagging and fouling tendency of Ind-coal than that of Phi-coal.

2.3. Flat-flame system for ash slagging

In this work, a novel particle sticking probability measurement system is developed based on the temperature-controlled flatflame burner, as shown in Fig. 4a. The flat-flame burner features a stable high-temperature environment for a rapid heating rate of entrained particles. Moreover, it provides the flexibility to control the ambient temperature, atmosphere and particle velocity (kinetic energy) independently by adjusting the flowrates of gaseous fuel, oxidizer and carrier gas [44]. Thus, it is able to well mimic the local condition of slagging in the boiler. For ash slagging, a removable corundum probe (high-purity >99%), 1.3 mm in diameter, was horizontally placed in the centerline, at a 6 cm height above the burner, as seen in Fig. 4a. This height is such chosen that the particle stream is sufficiently heated but does not diffuse. Meanwhile, the small probe size yields the Stokes number of injected particles as high as 30, indicating a near-unity impaction efficiency. The



Fig. 4. (a) Schematic of the ash slagging setup based on the flat-flame burner; (b) Photographs of the deposition processes; (c) Schematic diagram of the intersected area A_1 and the cross-sectional area of the feed tube A_2 .

corundum probe, both before and after ash deposition, was carefully weighted for quantification of the deposited mass.

Well-dispersed particles were fed from the 2-mm central stainless-steel tube by a specially-designed de-agglomeration feeder. More details of the feeder are referred to Ref. [44]. In this work, we used the chemically-stable, volatile free PSD and WWD as the entrained particles to eliminate the uncertainty of ash melting characteristics. The samples were grounded and sieved to 65-74 μ m in particle size. The carrier gas (N₂) flowrate was 0.3 L/min, and the total injected sample was 1.800-3.000 mg in one run lasting for \sim 5 min, as shown in Fig. 4b. It should be noted that after each run, the deposited probe was blown by compressed air (velocity \sim 50 m/s) to remove the loosely-packed fraction before the probe (with dense slag) was precisely weighted. For the optical observation of slagging dynamics, the impact-rebound/adhesion of particles at various temperature was captured by a high-speed camera (Phantom V311, 512 \times 512 pixels) at a frame rate of 3000 fps and an exposure time of 50 μ s.

A B-type thermocouple was used to monitor the flue gas temperature of the sampling point. Correction of radiation and conduction losses was performed both for the thermocouple and incident slag particles [44]. For the particle temperature, the emissivities of PSD and WWD are estimated as 0.80 ± 0.09 and 0.77 ± 0.08 , respectively, based on the model in Ref [45]. Combining the uncertainty of particle size (65–74 μ m), we derive an error of 1.6%–2.5% for particle temperatures. The uncooled probe has an estimated temperature ~300 K lower than the particle temperature after radiation loss corrections.

To quantify the sticking probability (η_{stick}) of impacting particles, we have:

$$\eta_{stick} = m_{stick} / (m_{feed} \cdot \eta_{impact} \cdot (A_1 / A_2)).$$
⁽¹⁾

Here m_{stick} is the mass of deposited particles (kg), m_{feed} is the mass of total particles fed in (kg), A_1 is the projected probe area intersected with the central tube, A_2 is the cross-sectional area of the central tube, and (A_1/A_2) characterizes the fraction of injected particles that reach the surface of the probe (see Fig. 4c).

The impaction efficiency η_{impact} is evaluated based on the effects of particle Stokes number (St) and flow Reynolds number (*Re*):

$$St = \rho_p d_p^{-2} U_0 / (9\mu_g d_c), \tag{2}$$

$$Re = U_0 d_c / v. \tag{3}$$

Here ρ_p is particle density (kg/m³), d_p is particle diameter (m), U_0 is characteristic flow velocity (m/s), μ_g is fluid dynamic viscosity (Pa•s), d_c is deposition probe diameter (=1.3 mm), and ν is the fluid kinematic viscosity (m²/s). By defining a shear Stokes number St_{γ} = 2.1832 St / (1 + 4.45 *Re*^{-1/2}) and *x* = log St_{γ}, the impaction efficiency reads as [25]:

$$\log\eta_{impact}/\log\eta_{0.1} = 1 - -g(x), \tag{4}$$

and

$$g(x) = \begin{cases} a_0 + a_1 x + a_2 x^{-1} + a_3 x^{-2}, & \text{it} x < -0.2, \\ \left[1 - b_1 (x + 0.79)^{-1} + b_2 (x + 0.79)^{-2} - b_3 (x + 0.79)^{-3}\right]^{-1}, & (5) \\ & \text{if} x > -0.2, \end{cases}$$

$$\eta_{0.1} = 1.0204 \times 10^{-3} \text{Re}^{-0.35054}.$$
(6)

Here $a_0 = 0.1844$, $a_1 = 0.2017$, $a_2 = 5.080 \times 10^{-2}$, $a_3 = 3.634 \times 10^{-2}$; $b_1 = 0.05119$, $b_2 = 0.2071$, $b_3 = 1.236 \times 10^{-3}$.

2.4. Sample characterization

The chemical compositions of raw coal ashes and deposits sampled in the boiler were determined by X-ray fluorescence (XRF) (SHIMADZU, XRF-1800). The crystallized mineral phases in coal ashes and deposits were identified by X-ray diffraction (XRD) (D8 advance, Bruker) with a scanning rate of 5°/min from 10° to 80°. For micromorphology characterization, we solidified the deposits into epoxy resin, then ground and polished to obtain a smooth cross-section for Scanning electron microscopy (SEM) (ZEISS Merlin Compact) - Energy dispersive X-ray detector (EDX) (Oxford) analysis. Ash fusion temperature (AFT) measurements of the coal ashes and deposits were carried out under an oxidizing atmosphere. This procedure involved heating an ash cone with a specific geometry at 15 °C/min up to 900 °C, and then changing to 5 °C/min. During this process, the initial deformational temperature (DT), softening temperature (ST), hemispherical temperature (HT), and flow temperature (FT) were recorded according to the specific shapes of the ash cones. Viscosity measurement was carried out by a Theta high temperature rotational viscometer (Theta-1700) under mild reducing atmosphere.

2.5. Thermodynamic equilibrium simulation

The high-temperature phase transition and melting properties of the coal ashes and deposits were studied by the thermodynamic equilibrium approach. In particular, *Factsage 7.1* with pure Substance (FactPS) and FACT oxide (FToxide) database was used to simulate the multiphase equilibrium, proportions of liquid and solid phase, as well as phase transitions at temperatures ranging from 800 °C to 1500 °C. The inputs of reactants were in forms of oxides according to the elemental content. The main gas phase



Fig. 5. Measured and calculated viscosities of PSD and WWD as a function of temperature.

Table 2						
Chemical	compositions	of	deposits	sampled	in	the
boiler						

Samples	WWD	PSD	FSD	HRD
SiO ₂	32.94	47.13	27.28	19.6
Al_2O_3	11.52	20.31	11.28	14.59
CaO	25.36	11.14	25.33	10.67
Na ₂ O	2.07	1.15	2.16	1.71
Fe_2O_3	18.16	14.27	15.83	29.79
MgO	6.66	2.97	7.36	3.25
SO_3	0.163	0.033	7.69	15.61
K ₂ O	0.70	0.89	0.74	1.11
TiO ₂	0.77	0.95	0.82	1.13
MnO	0.236	0.203	0.17	0.139
$R_{B/A}$	1.17	0.44	1.31	1.32

compositions involved in the simulations, i.e., CO_2 , O_2 and H_2O , were empirically defined to be consistent with the practical process, which accounted for 6.0, 2.3 and 0.98 wt% of total ash, respectively.

The particle viscosity (μ_p) of WWD and PSD was also evaluated by the Viscosity module in *Factsage 7.1* and further corrected via the solid fraction in the mineral system [46]. A good agreement was obtained between the calculated and the experimental results, at least within the experimental temperature range above 1290 °C, implying the reliability of viscosity calculation (see Fig. 5). Additionally, the viscosity of PSD is significantly higher than that of WWD at a temperature higher than 1300 °C, and both increase sharply after cooling to 1210 °C and 1165 °C, respectively.

3. Results and discussion

3.1. Physicochemical properties of deposits sampled from the boiler

Table 2 presents the compositions of four deposit samples, and Fig. 6 shows XRD results of the deposits. Specifically, the weight percentages of Si and Al in PSD are highest and mainly in the form of $CaAl_2Si_2O_8$ (anorthite), as seen in Fig. 6. The deposits downstream (FSD and HRD) contain fewer contents of silicon but are remarkably more abundant in sulfur content, which are mainly in the form of $CaSO_4$ (anhydrite), as indicated by the XRD results. Calcium shows a significant enrichment in WWD and FSD, and the dominant Ca-bearing minerals are $Ca(Mg,Al)(Si,Al)_2O_6$ (diopside), $Ca_2Al_2SiO_7$ (gehlenite) and $CaSO_4$ (anhydrite). Sodium contents in these deposits are close to Phi-coal ash, but are several



 $\label{eq:Fig. 6. XRD results of the deposits collected from the boiler. \\ A:CaAl_2Si_2O_8(Anorthite), D:Ca(Mg,Al)(Si,Al)_2O_6(Diopside), He:Fe_2O_3(Hematite), \\ Q:SiO_2(Quartz), An:CaSO_4(Anhydrite), N:NaAlSiO_4(Nepheline), \\ G:Ca_2Al_2SiO_7(Gehlenite). \\ \end{array}$



Fig. 7. Ash fusion temperatures of raw coal ashes and deposit samples.

times higher than that of Ind-coal ash, suggesting the strong slagging tendency of Na-bearing minerals. Fe₂O₃ (hematite) is abundant in all deposits, as verified by both XRD and XRF results. The distinct features of these samples clearly demonstrate the decisive role of temperature on the formation and deposition of mineral phases. In addition, the R_{B/A} of PSD is lowest among the deposits, while slagging onto the PS is much more severe than any other parts in the boiler. It indicates that slagging is so complex a process that it is difficult to be accurately described by the simple index of R_{B/A}.

Figure 7 shows the ash fusion properties of coal ashes and deposits. The ashes prepared from two raw coal samples are featured with similar DT and FT. However, the values of ST and HT differ greatly because of the discrepancy in the contents of the fluxing elements (mainly Na and K). On the other hand, the ash fusion temperatures of the deposits vary a lot.The FTs of WWD and FSD are even lower than the DTs of PSD and HRD. It can be possibly explained by the enrichment of Ca, Na and Mg in WWD and FSD with a disruptive effect on the network, leading to a decrease in the ash fusion temperature and viscosity [47]. On the contrary, the enriched Si and Al in PSD act as network formers and weaken the fusion propensity by the polymerization of Si-O tetrahedral network [48].



Fig. 8. Summary of slagging tendency of several ash samples, R_{B/A}: basic to acidic oxides ratio [21]; R_{Si/Al}: silica to alumina ratio [22]; R_{Si}: silica ratio [49]; A_F: ash fusibility [49].



Fig. 9. SEM-EDX analyses of PSD.

Figure 8 summarizes the slagging tendency of coal ashes and deposits predicted by several existing criteria, and the results are quite inconsistent. For instance, the Ind-coal ash is decided as highly prone to slag by $R_{B/A}$ and R_{Si} because of its extremely high content of iron, as seen in Table 1. On the contrary, low slagging tendency of Phi-coal based on the ash composition is concluded. However, the fusibility of Phi-coal ash tells a strong slagging propensity. These discrepancies, while pointing out the unreliability of such criteria, suggests the importance of elements activity and their heterogeneous spatial distribution regarding the slagging behaviors. It is especially important for low-rank coals rich in AAEM.

Figure 9 presents the SEM-EDX results of PSD. Two typical micro-morphologies of minerals can be clearly identified. The refractory bulky grains marked by 1# are enveloped by irregular, molten minerals (2#). EDX results show that 1# features a composition of Ca-Al-Si-O, while location 2# is lower in calcium content but more abundant in sodium, implying the presence of lowmelting-point sodium aluminosilicate. Figures 9b displays more details of the irregular region. It is noted that Al, Fe and O are 'concentrated' on the discrete particle surface, while Na, K and Si seem to be more 'dispersed' in the detected area, as evidenced from elemental mapping in Fig. 9b. The discrepancy is believed to vary the melting behavior of different mineral clusters. Besides, abundant grayish rod-like Fe₂O₃ crystals are embedded in the molten Na/Ca-aluminosilicate, as presented in Fig. 9c, which is in accordance with the XRD result in Fig. 6. Overall, the microscopic analyses further provide evidence on the enhancing effect of Na on forming low-melting-temperature eutectics and sticky slagging layers in coal combustion [12, 50, 51].

3.2. Thermodynamic phase transition of raw coal ashes and deposits

In this section, we performed a high-temperature thermodynamic equilibrium analysis for the raw coal ashes and two deposits based on their elemental compositions. The solid-liquid transition and the compositions of liquid phase are plotted in Fig. 10 as a function of ambience temperature for PSD and WWD. The shaded areas in Figs. 10a and 10d correspond to the temperature ranges of PS, FS and HR zones. For PSD (left panel), the measured FT (1384 °C) is close to the simulated liquid temperature (\sim 1350 °C), and with the temperature declining to the initial liquid temperature (1150 °C), the liquid slag solidifies dramatically. The simulated temperature range for PSD melting is consistent with the measured ash fusion temperatures, and coincides with the operating temperatures of the platen superheater zone. For WWD (right panel), the predicted solid-liquid transition still covers the range of the measured ash fusion temperatures. However, it is noted that at the FT (1225 °C) of WWD, the melt fraction is only \sim 50%. It can be interpreted by the much lower viscosity of WWD as compared with PSD for above 1200 °C, as shown in Fig. 5.

In the solid phase of PSD, $CaAl_2Si_2O_8$ and Fe_2O_3 are found to precipitate when the temperature drops below the liquid temperature, and are the dominant compositions. It can be confirmed by the XRD result in Fig. 6, which also validates the reliability of thermodynamic equilibrium analysis to understand the slag melting. Subsequently, NaAlSi₃O₈, KAlSi₃O₈ and magnesium silicate (the main chemical form of magnesium) appear successively and fast solidify at temperatures below 1200 °C. When the liquid phase disappears, the composition changes little with temperature. Notably, in the initial melting stage, the major elements are Si, Al, K and Na, indicating the fusibility of Na/K- aluminosilicate, as plotted in Fig. 10c. For WWD, Ca, Mg and Fe silicate or aluminosilicate are the dominant minerals in the range between 1300 °C and 1145 °C. Since then, NaAlSi₃O₈ and KAlSi₃O₈ crystallize quickly until 1100 °C, as shown in Fig. 10d.



Fig. 10. Thermodynamic equilibrium of mineral evolution, mineral phase transition and mass fraction of main elements in the liquid phase for PSD (a-c) and WWD (d-f) under different temperatures.

Since the sampled deposits result from the co-combustion of Phi-coal and Ind-coal, we further simulated the melting characteristics of raw coal ashes. As shown in Figs. 11a-11c, the liquid temperature of Phi-coal ash is higher than 1400 °C due to the presence of mullite. When the temperature drops below 1280 °C, CaAl₂Si₂O₈, Al₄Mg₂Si₅O₈ and Fe₂O₃ start to precipitate and reach constant contents rapidly. Meanwhile, NaAlSi₃O₈ and KAlSi₃O₈ contents increase progressively until 1015 °C (the initial liquid temperature). It is also manifested by the elemental compositions of the liquid slag in Fig. 11c, where Si, Al, Fe and Ca dominate at high temperatures above 1300 °C, while Na and K are the main basic species in the liquid slag from the initial melting stage to 1200 °C. By contrast, as depicted in Figs. 11d-11f, the initial liquid temperature (1170 °C) and liquid temperature (1480 °C) of Ind-coal ash are higher than those of Phi-coal ash. $CaAl_2Si_2O_8$ and Fe_2O_3 are the main mineral phases. A small quantity of Na/K- aluminosilicate starts to precipitate at 1360 °C and stabilizes as the temperature goes down to 1170 °C.

Recall the serious slagging on the panel superheater after cofiring the two coals. The deposit PSD consists mainly of CaAl₂Si₂O₈ and Fe₂O₃ (see Figs. 6, 10a and 10b), which is close to the Ind-coal results. It is no surprising because the blending ratio of Ind-coal (84.8 wt%) dominates. However, the excessive accumulation of PSD must be attributed to the greater melt fraction of Phi-coal ash. It can be deduced that Na and K compounds in Phi-coal ash, taking ~30 wt% of the total ash, contribute remarkably to the ash sticking propensity. Even worse, they are expected to strengthen the inner deposit/slag structure by the 'glue' effect [52], as verified by the SEM observation in Fig. 8.

Besides, it is noted $CaSO_4$ is formed by the both coal ashes at relatively low temperatures (<1050 °C), well explaining the accumulation of $CaSO_4$ in the FS and HR parts (see Fig. 6).

Figure 12 further compares the calculated melt fractions of raw coal ashes, PSD and WWD as a function of temperature. Reportedly, the reducing atmosphere facilitates the ash melting propensity by affecting the variable-valence iron species [40], but those iron-bearing silicates formed under reducing environment (e.g., olivine) are not identified in PSD, implying an oxidizing atmosphere in the PS region. The melt fraction of PSD largely lies between those of Phi-coal and Ind-coal ashes. In particular, the Phi-



Fig. 11. Thermodynamic equilibrium of mineral evolution, mineral phase transition and mass fraction of main elements in the liquid phase for Phi-coal ash (a-c) and Ind-coal ash (d-f) under different temperatures.

coal ash with high contents of Na and K melts at relatively low temperature and the melt fraction increases to 40 wt% at 1150 °C, at which PSD starts to melt. The sharp rise of the PSD melt fraction from 1150 °C to 1200 °C is attributed to the melting of feldspar (CaAl₂Si₂O₈, NaAlSi₃O₈ and KAlSi₃O₈) and Ca/Mg-silicates according to the thermodynamic calculation. The melting temperature of WWD is also lower than that of PSD because of the high proportion of AAEM. To summarize, the liquid phase is initiated by the fusion of low-melting-point Na/K-aluminosilicate, especially in Phi-coal ash. In contrast, Ca- and Fe-bearing minerals have higher melting temperatures. It well illustrates the discrepancies of different elements in promoting mineral melting.

3.3. Mechanistic studies on the sticking probability & AAEM contributions to slagging

To accurately evaluate the ash slagging propensity at different temperatures, we performed mechanistic experiments in the temperature-controlled flat-flame system. Details of the experi-

ments are described in Section 2.3. Figure 13 shows the measured sticking probability of the pulverized chemically-stable PSD and WWD. It is revealed that the sticking probability of PSD increases monotonously from 1200 °C to 1330 °C. Then the near-unity sticking probability above 1330 °C explains the severe slagging onto the platen superheater surface. Comparatively, the fusible WWD slagging occurs at lower a temperature (1080 °C) and quickly reaches the peak at ${\sim}1210$ °C. The videos filmed by high-speed camera in the supplementary material reveal more clearly the dynamic process of WWD particle impacting the probe. As shown Video 1#, almost all the impacting particles rebound at the room temperature. Whereas, the particles in Video 2# are partly captured by the probe with elevated particle temperatures around 1020 °C. When the particle temperature rises to 1230 °C or higher (Videos 3# and 4#), all impacting particles are found to stick and flatten on the probe surface.

Also shown in Fig. 13 are some model predictions, and it seems none of the existing models are fully consistent with the profiles of both samples. For the viscosity-based model [26], we use different



Fig. 12. Melt fraction of raw coal ashes, PSD and WWD as a function of temperature.

values of reference viscosity μ_{ref} to fit the experimental results. It turns out that the PSD result can be well fitted by $\mu_{ref} = 500$ Pa·s, but no μ_{ref} can fit the WWD data satisfactorily. Indeed, the value of μ_{ref} is not knowable *a priori*, and considering the failure for the WWD, the viscosity model predictions have presented defects, at least in our cases [29]. As an alternative, the linear melt fraction model overestimates the sticking probability of PSD between 1150 °C and 1300 °C, while distinctly underestimating the sticking propensity of WWD [30]. For the recently revised model that incorporates both particle kinetic energy and melt fraction [33], its predictions for both samples grow more slowly with increasing temperatures, as presented in Fig. 13.

Based on our experiments, we propose a nonlinear S-shaped, melt fraction (f)-based model for ash sticking probability, as shown in Fig. 14. It is quite reasonable when considering that the melt fraction from the thermodynamic equilibrium simulation has been validated by experiments in many aspects, as detailed in Section 3.2. Such a strategy was also adopted in the literature [29, 53]. The sticking probability is formulated as:

$$\eta_{stick} = \eta_{stick}^{max} - \left(\eta_{stick}^{max} - \eta_{stick}^{min}\right) \cdot \exp\left[-\left(\lambda f\right)^{\alpha}\right].$$
(7)



Fig. 14. Measured and predicted sticking probability of PSD and WWD as a function of melt fraction. The nonlinear model is the newly proposed Eq. (7). The linear melt fraction model [30] and kinetic energy-melt fraction model [33] are single-variable functions of the particle melt fraction and thus sample independent.

Here η_{stick}^{max} is the upper limit of the sticking probability (set as 1) and η_{stick}^{min} is the minimum value of the sticking probability taken at f = 0. The current model makes a compromise by introducing λ and α as sample-dependent, dimensionless constants. For PSD, the parameters are regressed as $\lambda = 1.4$ and $\alpha = 5.5$, while for WWD, $\lambda = 3.5$ and $\alpha = 3.1$. Clearly, it is seen from Fig. 14 that the new nonlinear correlation yields a more accurate sticking probability than the existing models. But we shall remark that the formula requires validation against more experimental data in future work.

We then elaborate the contribution of AAEM and Fe species on slagging. Figure 15a compares the predicted sticking probability of PSD, WWD and the raw coal ashes by the newly proposed Eq. (7). Furthermore, the contents of AAEM and Fe in the four samples are displayed in Fig. 15b. Whichever model parameters are used, Phicoal ash features a higher sticking probability than Ind-coal ash. This is a direct consequence of the greater melt fraction, as shown in Fig. 12. Thus, we may speculate that the severe slagging on the platen superheater surface is mainly ascribed to Phi-coal ash. By



Fig. 13. Measured and predicted sticking probability of PSD (a) and WWD (b) as a function of temperature. The experimental sticking probability is normalized by the maximum average value obtained in each case. For model predictions, the linear melt fraction model defines a linear increase of sticking probability with the particle *melt fraction* ranging from 10% to 70% [30]; the reference viscosity model determines the sticking probability by μ_{ref}/μ_p if $\mu_p > \mu_{ref}$, otherwise, it is one [26]; the critical viscosity model assumes the sticking probability to be zero if $\mu_p > \mu_{crit}$, otherwise it is one [31]; in the kinetic energy-melt fraction model, the sticking probability is an exponential correlation of the particle melt fraction, in which the excess energy ratio is considered [32].



Fig. 15. (a) Sticking probability of two slag samples and raw coal ashes predicted by Eq. (7); solid line: PSD parameters; dashed line: WWD parameters; (b) contents of AAEM and Fe in the coal ash and slag samples.

comparing the contents of Na+*K*, Ca+Mg, and Fe in Phi-coal ash and Ind-coal ash, it is found that the ash sticking propensity is well correlated with the amount of Na+*K*, regardless of Ca+Mg and Fe contents. It highlights the leading role of alkali metals in slagging. In contrast, Ind-coal has the highest Fe content but the least sticking propensity at the investigated temperature range. Bearing in mind that Fe mainly exists as discrete Fe₂O₃ (hematite), as exhibited in Figs. 6 and 8, it implies Fe is less effective than AAEM (Na/K and Ca/Mg) in forming slagging in the low-rank coal-fired boiler. Hence, the higher quantity of Na/K and Ca/Mg in WWD results in an even stronger sticking propensity than PSD.

4. Conclusions

This work comprehensively investigated the slagging mechanism of low-rank coal combustion. Deposits collected from a 660 MW wall-fired boiler were thoroughly characterized for compositions and phase transition at high temperatures. The sticking probability of chemically-stable slag particles was measured in a welldesigned temperature-controlled flat-flame system. The main conclusions are drawn as below:

- (1) For the first time, a flat-flame system under well-controlled conditions is developed for accurately quantifying the sticking probability of sampled pulverized deposits with thermally-stable compositions form the practical boiler. It also facilitates close optical observations of the dynamic slagging process. Few existing models can fully reproduce our measurements. A nonlinear formula is then proposed to correlate the sticking probability with the melt fraction of ash particles under high temperatures with sample-specific parameters.
- (2) Severe slagging takes place on the platen superheater surface in the investigated boiler cofiring the blends of two low rank coals (Phi-coal: Ind-coal = 15.2 wt%: 84.8 wt%). The compositions of fully-sintered deposit (PSD), including refractory anorthite (CaAl₂Si₂O₈), hematite (Fe₂O₃), are largely determined by the dominant Ind-coal ash. However, the excessive accumulation of PSD can only be attributed to the much stickier Phi-coal ash rich in low-melting-point Na-aluminosilicates.
- (3) Incorporating the sticking probability quantification and the elaborated sample characterization, we reveal a positive correlation of the ash sticking probability with the sodium con-

tent of the samples, almost regardless of the contents of alkaline earth metals and iron. It highlights the leading role alkali metals play in slagging. In contrast, iron exists as discrete hematite (Fe_2O_3) in the slag and is less effective in slag formation than AAEM.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.combustflame.2021. 111956.

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