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Effect of mineral components on sintering of ash particles at low temperature fouling conditions

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ABSTRACT

The ash deposition phenomenon in the coal gasification process is a severe problem for continuous operation. The ash deposition rate is influenced by the behavior of ash particles on the deposit target. Dropped coal ash particles, which exhibit different behaviors on the deposit surface such as attachment, rebounding, and removal, were observed through the view port of a drop tube furnace (DTF). The different behaviors may be influenced by the state of the ash particles such as the sintering phenomenon, which is a function of the mineral content. To verify the mineral components important in sintering among the ash particles, transformation of the particle shape of synthetic ash reacted at 900 °C was analyzed through a scanning electron microscope (SEM). The Fe, Ca, and Mg components were the main factors for sintering and agglomeration. The Fe, Ca, and Mg components sintered with Si and Al were observed in the mineral mapping of deposited coal ash by scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM–EDX) analysis. Sintering characteristics of coal ash were analyzed by the dilatometer technique. Sintering temperature, shrinkage, and sintering activation energy (E_{sin}) were all found to be related to the Fe, Ca, and Mg contents.

1. Introduction

Coal has continuously played a significant role in power generation and industrial sectors. The accumulation phenomenon of organic matter, corrosion products, inorganic particulates or other deposited matters on a heat transfer surface and the inner side of ducts/pipes is called fouling [1]. The deposition phenomenon in a thermo-chemical conversion process leads to operating problems of the plant [1–9]. Among deposited matter, the inorganic particulates play the key role in the fouling phenomenon. Alkaline earth and alkali metals are typically the main inorganic matter composing fouling accumulation. Alkali metals such as Na and K are vaporized while the coal particles are burned out. Fouling is first initiated by the vaporized materials, which provide a sticky surface on which to trap impacting particles. On the other hand, alkaline earth metals (Mg and Ca) and Fe compounds, which stick to the deposit surface due to inertial force, are exposed with the breaking up of char particles. They fuse and agglomerate with each other, after which big ash particles may be formed. Many researchers [10–13] have conducted to predict the ash formation with developed empirical indices about inorganic matter, such as the base to acid ratio, B/A (%Na_2O), total alkali contents, %Na_2O ratio in ash, and iron to calcium ratio. Although the empirical indices are well matched with their experimental results, these indices sometimes do not fit with the data from other research because of the limited number of coal samples tested. It is important to know what kinds of inorganic components influence the growth of fouling and particle agglomeration to develop empirical fouling indices and to predict fouling tendency. In general, if the surface temperature of the fouling layer becomes higher than a certain limit, sintering of the particles takes place [14]. Extracted inorganic matter from the fuel may be melted at lower temperatures than the inherent melting temperatures when forming eutectic compounds, which leads to minimization of the sintering temperature [15,16]. Sintering of particles leads to reduction of the void volume fraction, and reinforces particle contact force. The characteristics of the sintered layer and powdery layer influence particle movement because of their different particle stickiness. This affects the growth of fouling. Sintering of ash powder has been widely researched. The viscous flow mechanism, which indicates that an amorphous material possesses temperature-dependent viscosity, was reported by Raask [17] to be an important mechanism in ash sintering. Frenkel [18], who developed the model for the viscous flow mechanism, was one of the first researchers to study particle sintering. The author suggested that crystalline materials have similar characteristics to amorphous materials under the effects of surface energy. The variation of surface energy influences the reduction of volume between clusters of packed spherical particles of the same size. Consequently, the physical dimensions of the particles change because of sintering occurring between the particles. In general, the variation of particle characteristics with temperature difference, such as particle sintering, is measured by using dilatometry techniques. The sintering characteristics of deposited ash particles may influence the fouling rate. Furthermore, ash particle movement

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Table 1
Basic analysis of coal samples in the experiment.

		Centennial	Taldinsky	WH	Zhongmei	Bengalla	LG	Cyprus	MSJ	Kideco
Proximate analysis	V.M.	30.05	29.58	32.13	26.77	36.72	53.38	46.64	51.29	50.10
(dry-basis, wt.%)	F.C.	53.48	56.73	48.67	64.15	44.76	38.21	47.30	42.82	48.23
	Ash	16.47	13.69	19.20	9.42	18.51	8.42	6.06	5.91	1.67
Ultimate analysis	C	83.04	88.62	88.18	82.91	83.18	76.59	68.22	75.41	64.12
(ash-free basis, wt.%)	H	5.16	5.30	5.70	4.05	5.41	5.38	5.25	5.34	5.20
	O	9.56	2.98	3.05	11.26	8.64	15.25	24.55	15.74	29.91
	N	1.69	2.54	1.95	0.74	1.91	1.55	1.19	1.78	0.24
	S	0.55	0.68	1.32	1.04	1.09	1.05	0.79	1.46	0.53
Heating value (HHV)	MJ/kg	27.36	29.60	27.75	28.83	25.81	27.38	28.56	28.52	28.17

such as attachment, rebounding and removal on deposit target is affected by the velocity of the impacting particles [19,20] as well as the sintering rate by the chemical components of the ash.

In this study, it was investigated which inorganic matter plays the key role in fouling growth and agglomeration. To examine the behavior and characteristics of ash particles, experiments were conducted in a drop tube furnace (DTF), in which the behavior of coal ash particles under gasification conditions can be investigated experimentally. Pulverized coal samples were injected into the DTF under gasification conditions, and the ash particles were deposited onto the sample collector. The behavior of the ash was filmed through the view port near the sample collector. The basic characteristics of ash particle movement on deposit surface were examined. Deposited ash samples were analyzed by mineral mapping of scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM–EDX) for verification of the mineral distribution in the fouling agglomeration. Synthetic ashes, which are general chemical reagents such as chloride compounds of Fe, Ca, Mg, Na, and K, were used for determining the agglomeration shape of particles according to the mineral components present. To verify the effects of coal mineral components on sintering behavior, the sintering characteristics of various bituminous and sub-bituminous coal ashes were confirmed by measuring the variation of physical dimensions through the dilatometer analysis.

2. Experimental section

2.1. Sample preparation

Nine different coal samples were chosen for the experiments, which represented a wide range of mineral contents of bituminous and sub-bituminous coal. The basic properties of the coal samples measured by proximate and ultimate analyses are illustrated in Table 1. The inorganic chemicals of ash are analyzed by X-ray fluorescence (XRF). The chemical compositions and fusion temperatures of the coal ash samples are also given in Table 2, in which the chemical compounds, SiO_2, Al_2O_3, Fe_2O_3, CaO, and MgO were assigned as the major components of the ash. The deposition behavior of the coal ash was studied using the DTF adopted to view port, as shown in Fig. 1. In the case of synthetic ash, the ash was composed of general chemical reagents. They were chosen as the main acid, alkali and alkaline-earth metals in ash components such as Si, Al, Fe, Ca, Mg, Na, and K. Excluding the acid metals, the chosen original chemicals of alkali and alkaline-earth were from chloride metal compounds to verify their agglomeration behavior under low ash fouling temperatures. The individual melting points of the synthetic ashes used in the experiment are listed in Table 3. The original coal samples, as well as the synthetic ash, were dried and crushed in a fan-type disk mill, and then separated under 150 μm. All of the particle samples were dried in an electric oven at 60 °C for 24 h before conducting the experiments.

2.2. Experimental facility and method

2.2.1. Drop tube furnace for coal ash behavior

The DTF is able to simulate the conditions of a coal gasifier. The system mainly consisted of a coal sample injector, pre-heater, main tube furnace reactor and deposit probe. The temperature of the main reactor was set to 1300 °C. The temperature zone was divided into 3 zones in order to easily control the reactor temperature by means of a proportional–integral–derivative (PID) system. Pulverized coal was fed at the top of the DTF using a screw feeder with a flow of 2 l/min nitrogen gas (primary gas), and the injected coal particles then reacted with the concentric oxygen gas (secondary gas) in the DTF. During each experiment, the coal sample was continuously fed into the DTF for 10 min at a rate of 0.5 g/min, and the molar ratio of O_2/coal was set to 0.9.

The pre-heater was set to 1000 °C to prevent sudden thermal expansion of the flue gas, including the coal. The coal particles were transformed through devolatilization and char burning before ash formation, and were then collected on the ash collector of a deposit probe. The deposit probe was inserted vertically into the end of the lowest furnace section. The surface temperature of the probe was set to 700 °C during the experiment. The DTF experiment was started at the desired temperature, and the ash deposition behavior was filmed through the external view port near the probe. After the experiment, deposited ash was analyzed through scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX).

2.2.2. Electric muffle furnace for synthetic ash

To certify the critical alkali and alkaline-earth metals in particle agglomeration and fouling growth, agglomeration tests of synthetic ash were conducted by using an electric muffle furnace. Table 3 shows the

Table 2
Chemical components and fusion temperatures of the ash from coal samples.

		Centennial	Taldinsky	WH	Zhongmei	Bengalla	LG	Cyprus	MSJ	Kideco
Inorganic analysis (wt.%)	SiO_2	62.1	60.51	55.44	60.08	58.73	49.94	62.29	44.33	42.85
	Al_2O_3	15.5	27.59	31.39	25.74	27.86	28.66	16.89	28.66	20.53
	Fe_2O_3	5.89	5.14	4.44	7.44	5.85	8.56	7.19	11.21	18.12
	CaO	1.69	2.26	5.24	3.78	4.85	5.41	8.34	6.94	12.31
	MgO	0.07	1.44	0.71	0.49	1.21	2.83	2.16	3.15	2.84
	Na_2O	0.03	0.46	0.29	0.57	0.49	1.42	1.10	2.78	0.87
	K_2O	8.49	1.79	0.71	1.03	0.68	2.01	1.10	1.90	1.55
	TiO_2	6.3	0.75	1.53	0.87	0.68	0.97	0.93	0.98	0.93
Ash fusion temp. (°C)	Fluid	>1550	1450	1412	1421	1356	1388	1289	1310	1408