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Exploration of effective bed material for use as slagging/agglomeration preventatives in circulating fluidized bed gasification of high-sodium lignite

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ABSTRACT

An effective bed material is crucial to remit slagging/agglomeration for high-sodium lignite during circulating fluidized bed (CFB) gasification. In this study, the exploration of effective bed material for use as slagging/agglomeration preventatives of high-sodium lignite mined from Zhundong district was conducted in a 0.4 T/D CFB test system. Three mineral materials, SiO_2-based quartz sand, Al_2O_3-based corundum and CaO/Fe_2O_3-rich boiler ash collected from bottom ash of a CFB industrial boiler, were selected as the potential bed material. Their impacts on the de-fluidization tendency, transformation and migration of alkali metals, priority of reactions involving sodium-based species, formation of liquid slags and ash fusibility were investigated through the characterization of ash samples and thermodynamic equilibrium calculation by Factsage 6.1. The particle-size analysis results presented the improvement of three bed materials in the growth of ash particles followed the order of quartz sand > corundum > boiler ash. This order was ascribed to their reaction priority with sodium-based compounds in coal and the chemical property (mainly referring to ash fusibility) of corresponding products. Generally, reactions between sodium-based species and minerals (Al_2O_3 and SiO_2) were the main way of sodium retention within the ash in gasifier. As boiler ash was used, however, Al_2O_3 and SiO_2 preferentially reacted with the CaO enriched in boiler ash rather than sodium-based species in coal, resulting in very low sodium retention. Under such circumstances, computing results suggested the ash in gasifier could withstand higher gasification temperature, consistent with the measured results of ash fusibility. Additionally, the gasification temperature operating range could be largely widened by simply increasing CaO/Fe_2O_3 content in gasified ash. Therefore, boiler ash is an effective and low-cost bed material for use as the slagging/agglomeration preventative of high-sodium lignite during CFB gasification.

1. Introduction

As a clean coal utilization technology, gasification has been widely used in processing coal, especially for low-rank coal (lignite and sub-bituminous coal) that accounts for over half of the total coal reserve in China [1,2]. The industrial gas produced via coal gasification is a low-cost gas fuel for industrial stoves in the heat, steel, metallurgy, petrochemicals and chemical industries. Since the conventional fixed bed gasification technology is now under pressures of capacity, cost and pollution, the burgeoning fluidized bed (FB) gasification technology gradually occupy the market because of its widespread adaptability of fuels, high gas yield and little environmental pollution [3]. As one of the promising coal gasification technologies, circulating fluidized bed (CFB) gasification technology has been extensively studied and applied to low-rank coal [4–7].

However, when the high-sodium lignite (namely Zhundong coal) exploited in Northwest China and estimated with 390 Gt reserve [8], was used as the gasification fuel in CFB, severe ash-related problems such as agglomeration and slagging occurred [9–11]. To date, extensive researches have confirmed the too high sodium content in coal ash as the chief reason [12–14]. As reported, alkali metal, earth-alkaline metal oxides and iron oxide were reported as well-known catalysts for coal gasification [15–18]. If their contents are too high at high temperature, however, some of them would be trapped in silicates and aluminosilicates [19,20] and then loss the catalytic capacity. The trapped alkaline, especially silicates, are always characterized with low melting points (900–1100 °C) [13] and high viscosity, which signifies that the gasifier is threatened with slagging and agglomeration. Despite the availability of free ash-related problems during operation [21], a low gasification temperature is still not suggested due to the low heating value gas and low efficiency (cold gas efficiency and gas yield) [22]. Therefore, slagging and agglomeration have become a troublesome issue for

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utilization of the special high-sodium lignite in CFB gasifier.

Recently, some solutions have been introduced to realize the utilization of high-sodium low-rank coal. Blending with other fuels is one common way to control the above-mentioned issues. Wang et al. [23] found serious ash deposition still existed when co-firing ratio of Zhundong coal was about 60% in a 350 MW tangential-fired boiler after continuous operating for six months. This study also pointed out the important role of sodium and calcium sulfates on the slagging and ash depositing on convection heating surfaces. Jiang et al. [24] carried out the chemical looping co-combustion of sewage sludge and Zhundong coal with natural hematite as the oxygen carrier. During the co-combustion process, no ash agglomeration was observed due to the generation of high-melting-point sodium aluminosilicates and sodium pyrophosphate aluminum. Although the co-fired ash was deposited on the oxygen carrier, its reactivity was barely affected, which indicates the hematite oxygen carrier possessed good long-term reactivity. Besides, co-pyrolysis of Zhundong lignite and pine sawdust [25], and co-combustion of Zhundong coal and oil shale/Australian bituminous [26,27] were reported. However, most of the above work failed to solve these issues, even directly ignored them without any discussion, or had to accept a poor economy.

Bed materials and additives also play an important role in slagging, agglomeration and fouling during the thermal conversion of high alkali metal coal [28–37]. Generally, the minerals rich in both alumina and silica were found most effective to retain sodium within the coal ash for use as fouling preventatives, while the retained sodium had to exist in the high-melting-point form in order to avoid slagging and agglomeration in furnace. Moreover, the common low-priced minerals or industrial waste used as bed materials and additives will greatly reduce the cost and increase the possibility of large-scale application. As coal ash performs a stronger sintering tendency in reducing atmosphere [38], more efforts should be put to coal gasification or pyrolysis, which was unfortunately ignored by recent studies.

In this study, an appropriate bed material is considered as the breakthrough in solving slagging and agglomeration. Three minerals were selected as the potential bed material for gasification of a high-sodium lignite in a 0.4 T/D CFB test system, two of which are common SiO_2-based and Al_2O_3-based minerals and the other one of which is the screened waste ash from the slag bleed hole in a CFB industrial boiler. Apart from analyzing the collected ash by some characterization methods, the thermodynamic equilibrium calculation was employed as auxiliary means to explore the feasibility of the three potential bed materials. The aim of our works is to find an effective and low-cost bed material for use as the slagging/agglomeration preventative of high-sodium lignite during CFB gasification.

2. Experimental section

2.1. Fuel and bed material

Zhundong coal mined from the district of Tianchimulei was used in this study, and named as TCMLc here. As seen in Table 1, the acidic oxide content in ash is very low, such as 3.73% for SiO_2, 6.16% for Al_2O_3, and 0.41% for TiO_2, while alkaline oxides including CaO, Fe_2O_3, MgO, Na_2O and K_2O are rich, especially Na_2O with the high content of 7.28%. The too high alkaline -acidic ratio indicates the used coal has a strong slagging tendency.

Three mineral materials, namely BM-1, BM-2 and BM-3, were used as bed materials, and their chemical components are shown in Table 2. Please note, quartz sand and corundum, which are separately described as BM-1 and BM-2 in this paper, are common SiO_2-based and Al_2O_3-based bed materials, of which corresponding contents are up to 95.64% and 90.05%. The bed material of BM-3 is the screened bottom ash from a CFB industrial boiler. Due to the desulfurization in furnace for the CFB industrial boiler through adding limestone, CaO content in BM-3 is up to 61.36%, and mainly exists in forms of lime (CaO) and limestone ($CaCO_3$) via XRD analysis. Additionally, Fe_2O_3 content in BM-3 is also high, ~22.42%, which was related to the coal used in the CFB industrial boiler.

Table 1
Properties of TCMLc.

Proximate analysis (%, air dry basis)	
Fixed carbon	55.48
Volatile	27.02
Ash	3.16
Water	14.34
Ultimate analysis (%, dry basis)	
C	75.34
H	3.53
O	16.31
N	0.61
S	0.53
Cl	0.065
Ash composition (%)	
SiO_2	3.73
Al_2O_3	6.16
Fe_2O_3	5.37
CaO	33.45
MgO	5.42
TiO_2	0.41
SO_3	29.34
K_2O	0.45
Na_2O	7.28
Others	8.39

To ensure normal fluidization of coal and bed material particles, the used coal and bed materials were crushed and screened in a size range of 0.1–1.0 mm and 0.18–0.71 mm, respectively.

2.2. Test system

Gasification experiments of TCMLc were conducted in a 0.4 T/D CFB test system, of which schematic diagram is shown in Fig. 1. As a CFB, this test system is primarily composed of a riser with the height of 4.4 m and the inner diameter of 150 mm, a cyclone and a loop seal, which is also equipped with air supply system, auxiliary heating system, water cooling system, feeding coal system and data acquisition system, etc.

Sampling positions (P1–P16) were designed along the gas flow direction. In this study, bottom ash (P1) and fly ash (P4) were collected and focused in order to investigate the slagging/agglomeration tendency in the main circulation loop. The two ash samples were collected under stable working condition through sampling cans by opening or closing its valve.

2.3. Test conditions

Three mineral materials were separately tested as the bed material in a single experiment. In each test, the temperature measured in T1 was considered as bed temperature. As seen in Table 3, some differences among the three tests in operational parameters (bed temperature and reaction atmosphere) occurred under stable working condition. Here, these differences are not considered influential to the purpose of comparison among the three tests.

2.4. Sample characterization and calculation

A series of characterization methods were employed to analyse ash samples. Particle-size distribution of bottom ash was determined through sieve analysis. Additionally, chemical properties of bottom ash and fly ash were measured by an X-ray diffractometer (XRD, Panalytical, Netherlands) and an X-ray fluorescence (XRF, XRF-1800, Japan) in order to determine their crystalline phases and chemical components, respectively.