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Full Length Article

A study on fragmentation behavior, inorganic melt phase formation, and carbon loss during high temperature gasification of mineral matter rich fraction of Pittsburgh No. 8 coal

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HIGHLIGHTS

- Ash fragmentation to finer size is limited irrespective of particle sizes.
- Mineral matter rich fraction forms melt phase at lower conversion (<30%).
- Encapsulation of organic matter by melt phase leads to unburnt carbon for both the particle sizes.

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ABSTRACT

Computational models are being developed to determine the fate of organic and inorganic matter during gasification to improve operational reliability and carbon conversion in entrained flow gasifiers. Current models being developed to simulate entrained flow gasifiers assume homogeneity of the coal phase. However, coal is not homogeneous, and various size and density fractions behave differently during gasification. Therefore, a more accurate model can be developed by determining the behavior of various size and density fractions of coal. From that perspective, two particle size distributions (75–106 μm and 212–425 μm) of the mineral matter rich fraction of Pittsburgh No. 8 coal obtained using the float and sink method were gasified in a laboratory-scale entrained flow reactor at 1773 K. Char-ash particles from the two size fractions were characterized using Scanning Electron Microscope-Energy Dispersive X-ray Spectroscopy (SEM-EDS), X-ray Diffraction (XRD), Mossbauer spectroscopy, thermogravimetric analysis, and a micro-sifter for particle size distribution (PSD). The char-ash sample derived from the coarser-sized fraction (212–425 μm) showed limited fragmentation (~8%), while the finer-sized fraction showed no fragmentation. On the other hand, agglomerates were seen for both the particle size fractions with the finer-sized fraction having more melt phase and agglomerates than the coarser-sized fraction. The melt phase composition in the agglomerates is linked to the presence Fe^{2+}-glass and fayalite for both the particle size fractions. The data also confirm that the mineral matter rich particles form melt phase and initiate slagging in the gasifier even at a lower conversion level (<30%). The melt phase thus formed covers the carbonaceous matter in the particle and contributes substantially to the unburnt carbon in the slag.

1. Introduction

IGCC technology employing pressurized entrained flow gasifiers is considered one of the most efficient ways of generating electricity by reducing coal's environmental footprint. However, high overall cost and poor reliability are among several factors that make the IGCC system less favorable [1]. Issues like syngas cooler fouling, slag mobility and slag discharge, and unburnt carbon in the slag affect the overall efficiency and operational reliability of the gasifier [1–3]. Addressing these issues requires a deeper understanding of the transformations of mineral matter during gasification.

Transformations of mineral matter at high temperatures are complicated due to the heterogeneity of inorganic species in terms of their composition and association in coal. Inorganic particles in coal can occur as discrete particles with little or no association of organic matter (i.e., excluded mineral matter) or can be intimately

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mixed with organic matter (i.e., included mineral matter). This difference in association of inorganic species in coal can affect slagging (i.e., deposition in the radiative section of the gasifier) and fouling (i.e., deposition in the convective section of the gasifier) behavior. Wall et al., based on a study conducted in a pilot-scale oxygen-blown Hitachi gasifier, reported that high-density inorganic-rich particles in coal, particularly pyrite, tended to be tapped as slag [4]. Another study conducted in a bench-scale OMB gasifier using a Chinese bituminous coal concluded that the slag formed near the burner plane and gasifier dome comprised inorganics mainly derived from excluded mineral matter [5]. A computational model developed by Weber et al. for predicting ash deposition in a pulverized coal fired boiler also showed that the number of particles impacting the wall surface increases with an increase in particle size [6]. Although the extent of inorganic species captured on the gasifier wall surface is also dependent on factors like particle velocity, particle temperature, and the composition of the particles [7,8], it is imperative that the ash particle size distribution is known for accurate prediction of ash deposition in various sections of the gasifier. Ash fragmentation is known to alter particle size distribution. Previous studies conducted in combustion/pyrolysis conditions have noted that the fragmentation was not significant among particles rich in mineral matter [9,10]. But these studies were restricted to particle size in the pulverized coal range. By including ash fragmentation behavior for coarser sized fraction typical of the PSD to slurry fed gasifier, the accuracy of the ash deposition model can be improved. This in turn would be useful in predicting char conversion and the overall efficiency of the gasifier.

Mineral matter transformations can also affect the fate of organic matter associated with it. There are number of studies that reported existence of unburnt carbon in the slag collected from industrial gasifiers [11–13]. Wu et al. reported that carbon in the coarse slag generated from a commercial gasifier can be up to 35% [11]. Montagnaro et al. also observed the carbonaceous phase in the coarse slag generated from a commercial gasifier and concluded it was a result of entrapment of organic matter by the slag [12]. The contribution of carbonaceous matter in the mineral matter rich fraction towards unburnt carbon slag is unclear from these studies. Li et al. performed ash deposition experiments using Illinois No. 6 coal under gasification conditions using a laminar flow reactor and deposition probe. Based on the study, the authors observed that the particle stickiness on the probe increased dramatically above critical conversion. The dramatic increase in particle stickiness above the critical conversion was suggested to be the result of particle surface covered by molten phase generated from included mineral matter. The authors also suggested that excluded mineral matter contributed to deposits on the probe below the critical conversion [14]. However, the authors did not provide the proportion of included and excluded mineral matter in the feed. To the knowledge of authors, there is not a single gasification study conducted under controlled conditions that determined the melt phase formation and the fate of organic matter associated with mineral matter rich fraction representing finer and coarser-sized fractions of the PSD typical of slurry fed gasifiers. Analyzing the behavior of mineral matter rich fraction for melt phase formation and its consequent effect on unburnt carbon can be useful in developing better predictive models.

The objectives of the study were to determine fragmentation behavior, and melt phase formation of coarser- and finer-sized mineral matter rich particles representing the PSD of feed typically used in slurry-fed gasifiers at high heating rates (>1 $\times$ 10^4 K/s) and high temperature (1773 K) and their consequent effect on unburnt carbon. Two particle sizes of a mineral matter rich fraction were obtained from Pittsburgh No. 8 coal using the gravity separation (float-sink) method. These particle size fractions were gasified in a laboratory-scale entrained flow reactor at The Pennsylvania State University. The details of the reactor are described elsewhere [15]. The char/ash samples were obtained at a residence of ~0.65 s in the reactor. Particle size, composition, and morphology of char-ash particles collected were determined by a variety of techniques to determine the organic and inorganic transformations during gasification.

2. Experimental

2.1. Samples

The mineral matter rich fraction examined in this study was obtained from Pittsburgh No. 8 bituminous coal. The coal was dry ground in an industrial rod mill with a PSD very similar to that of coal fed into a commercial slurry-fed pressurized entrained flow gasifier. A representative sample of the ground parent coal was then sieved to obtain two particle size (PS) fractions: PS6 (75–106 μm) and PS3 (212–425 μm). These two particle size fractions were chosen as they cover ~40% of the coal fed to a commercial slurry-based entrained flow gasifier. Each particle size was further separated into two mineral matter rich fractions, SG3 (1.6–2.6 g/cc) and SG4 (>2.6 g/cc), by the float-sink method using organic liquid mixtures of different densities (toluene/perchloroethylene and 1,1,2,2, tetrabromoethane/perchloroethylene). A total of four mineral matter rich fractions were obtained. The mass of SG4 fraction was ≤0.3 wt% of whole coal for both the particle sizes. Therefore, only SG3 fraction was used in this study. The SG3 fraction constitutes about 30% of the mineral matter in coal. The properties of the samples (SG3PS3 and SG3PS6) are given in Table 1. The inorganic composition presented in Table 1 was based on X-ray diffraction, computer controlled scanning electron microscopy (CCSEM) analysis, and Mossbauer analysis of the SG3 fraction of the whole coal sample reported elsewhere [16].

2.2. Preparation of char-ash samples

All the experiments were conducted in an electrically heated entrained flow reactor operated under gasification conditions. A sample of about 30–40 g was fed into the reactor at a stable feed rate. The feed rates of coarser- and finer-sized fractions were 3.59 ± 0.02 g/min and 3.10 ± 0.02 g/min, respectively. The gas temperature in the isothermal zone was maintained at 1773 ± 12 K.

Table 1
Properties of samples studied.

Sample	SG3PS3	SG3PS6
Particle size	212–425 μm	75–106 μm
Mass yield from whole coal (air dry basis)	4 wt%	4 wt%
ρ_{He} (g/cc)	1.65	1.67
Proximate analysis (dry basis, wt%)		
Volatile matter	15.8	16.0
Fixed carbon	21.8	18.7
Ash	62.4	65.3
Ultimate analysis (dry basis, wt%)		
C	29.0	23.9
H	2.1	1.6
N	0.5	0.4
S	5.0	8.3
O[a]	0.9	0.4
Inorganic composition[b]	Pyrite, Pyrrhotite, Calcite, Illite/Muscovite, Kaolinite, Gypsum, Jarosite, Quartz, Rutile, Montmorillonite	

[a] By difference.
[b] Based on data from CCSEM, XRD and Mossbauer spectroscopy.