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Characteristics of solid by-products from entrained flow gasification of Australian coals

Alexander Y. Ilyushechkin *, Daniel G. Roberts, David J. Harris

CSIRO Energy Technology, PO Box 883, Kenmore, QLD, 4069, Australia

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ABSTRACT

Coal mineral matter undergoes a series of transformations during entrained flow gasification before exiting the gasifier as liquid slag, fly ash, or fly slag. As a result of these transformations, there is the potential for the composition of the slag and fly ash to differ significantly from that of the mineral matter in the feed coal.

This work presents new data from the testing of four Australian coals in a 5 MW_{th} pilot-scale entrained flow gasifier and describes analyses of the morphology, microstructure, and composition of samples of solid by-products obtained from the pilot-scale test program. The results of these analyses indicate some significant differences in composition between the mineral matter of the feed coal and the solid slag and fly ash by-products, which also impacts on the viscosity behaviour of the slags. These differences are primarily attributable to two factors: the partitioning of the mineral matter between fly ash and slag in the gasifier and, in some instances, the interaction of the fresh slag with slag produced from different coals and/or test runs which was already present on the wall of the gasifier.

1. Introduction

It is becoming increasingly important that efficient and environmentally sustainable technologies are available for deployment in energy production. Coal-based power stations generate more than 40% of the world's electricity, and the proportion of coal-based power is even greater in the fastest growing economies, such as China and India [1]. The continued use of coal for power generation will require new technologies, such as Integrated Gasification-Combined Cycle (IGCC) to increase generation efficiency and decrease emissions of greenhouse gases. IGCC technology combines the advantages of a relatively cheap fuel (coal) with the efficiency, economics, and environmental performance of gas turbines, whereas advanced IGCC systems incorporating new CO shift, and CO_2/H_2 separation technologies have the potential to produce H_2-based power with integrated capture and sequestration of CO_2.

All of the current IGCC demonstration power plants are based on entrained-flow gasification, which allows high throughput combined with fuel feedstock flexibility. The entrained flow gasifiers are slagging gasifiers, requiring the mineral matter in the feed to melt and flow out of the gasifier as a molten slag. A number of laboratory studies have focussed on the impact of coal properties on gasification performance, with particular emphasis on the key processes associated with slag formation and flow and the conversion behaviour of coal to syngas via gasification reactions [2–9]. Research at larger scales using entrained flow reactors [10,11] and gasification facilities [12,13] has generated data that allow us to interpret overall coal gasification performance under practical gasification conditions using fundamental coal conversion and slag formation characteristics.

Slagging characteristics are important for continuous operation in an entrained flow gasifier. Gasifiers lined with water-cooled walls require these walls to be covered by a layer of solid (crystallised) slag, over which newly forming liquid slag will form and flow [14]. Refractory-lined gasifiers have no requirements for minimum slag layer formation; however, even in refractory lined gasifiers, a suitable frozen layer is an important barrier preventing undesirable interactions between the slag and the refractory lining which can lead to corrosion or erosion problems. Wall-slag formation and continuous slag taping depends on the operating temperature and slag composition which resulted by coal mineral matter transformations in gasifier.

Not all of the mineral matter in the feed coal goes to liquid slag that forms on the wall of the gasifier and flows through the slag tap to be collected in the slag hopper. Small particles of mineral matter that have not melted and some small particles of molten slag will be carried out of the gasifier by the product gas and report as fly ash, which in a partial or complete quench arrangement is largely collected along with unburnt carbon as the solid component of the gas quench waste water.

Due to the potential for partial melting of the mineral matter and the entrainment of molten particles in the gas phase, the routes by which coal-derived mineral matter pass through the gasifier include a series of separation and combination processes (Fig. 1). The interaction of these processes results in the formation of a range of mineral-matter-

* Corresponding author. Tel.: +61 7 3327 4187; fax: +61 7 3327 4455.
E-mail address: alex.ilyushechkin@csiro.au (A.Y. Ilyushechkin).

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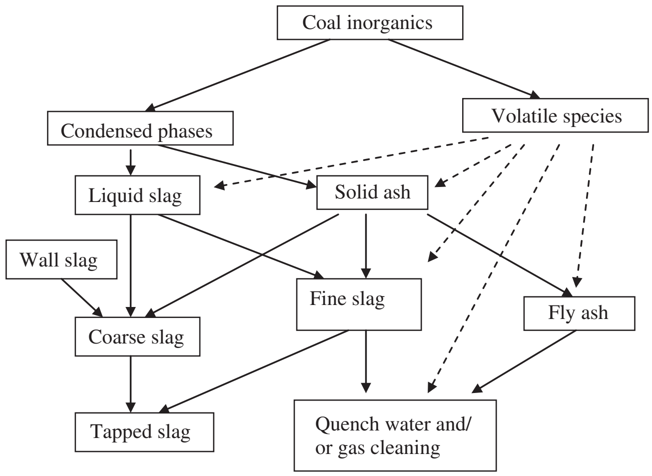

Fig. 1. Schematic illustration of mineral matter transformation in entrained flow gasification.

derived solids: coarse and fine slag collected in the slag tapping system, and fly ash collected by the quench water (in a gas-quench gasifier) or in the syngas particulate removal (cleaning) systems.

The fusibility of coal mineral matter and slag flow properties depend on their composition and can usually be broadly interpreted using the position of these bulk compositions and liquidus temperatures, in the quaternary SiO_2-Al_2O_3-CaO-FeO_x system (as major components in coal slags) [4,9,15,16]. It is expected that the partitioning of mineral matter to discrete process streams in a gasifier, and the effects of enrichment and depletion of various species in the mineral matter composition in the different process streams would have impacts on the viscosity behaviour of the bulk of the slag formed [15]. The composition of "real" slag would be dependent on the gasification process conditions and potentially be quite different to the composition of ash or slag prepared from the same coal under laboratory conditions and subsequently used for modelling or experimental investigations into slag formation and flow. This is likely to complicate interpretation of results and therefore the assessment of coals for use in entrained flow gasification.

In this work, the characteristics of slag and fly ash are studied using samples collected from a 5 MW_{th} pilot-scale entrained flow gasifier operated with a partial syngas quench. The composition of tapped slags and solids collected from the quench water are investigated and compared with the mineral matter compositions of the feed coals. Viscosity and flow behaviour of the tapped slags are compared with measurements using slags made from laboratory-produced ash from the same parent coal, and differences are interpreted with respect to observed compositional variations between solid phases and the potential impact on coal assessment for use in entrained flow gasification.

2. Experimental

2.1. Coal samples

Four Australian thermal coals were used in this work—they are designated here as CRC701, CRC702, CRC703 and CRC704. These coals were chosen to cover a wide range of coal types and to provide where possible a degree of consistency with the extensive research programs previously undertaken by the authors and their colleagues [2,3,8–10]. The same parent coals were used for both the laboratory and pilot-scale test program. Proximate and ultimate analyses of these parent coals are listed in Table 1. CRC701 is a sub-bituminous coal from Western Australia, CRC702 is a high volatile bituminous coal from the Hunter Valley in New South Wales, CRC703 is a semi-anthracite from the Bowen Basin in Queensland, and CRC704 is a sub-bituminous coal from the Surat Basin in Queensland. The high ash fusion temperature of CRC703 required it to be fluxed at a rate of 30 kg limestone to 1000 kg coal [13]. The fluxed sample is identified in this work as CRC703f.

Table 1
Characteristics of coals used in the trials campaign. Analyses are "as prepared" for the tests (ar = as received, daf = dry ash free, and HHV = higher heating value).

	CRC701	CRC702	CRC703f	CRC704
Proximate analysis, % as received				
Moisture	6.4	1.3	0.9	2.3
Ash	5.6	8.7	9.6	11.7
Volatile matter	36.2	34.4	7.2	44.1
Fixed carbon	51.8	55.6	82.3	41.9
Ultimate analysis, % dry, ash-free				
C	76.1	86.3	93.5	79.0
H	4.5	5.2	4.0	5.8
N	1.5	1.9	1.9	1.0
S	1.1	1.2	0.7	0.3
O	16.8	5.3	0.0	14.1
AFT fluid (reducing), °C	1290	1310	1430	1440
Specific Energy (HHV), MJ/kg as received	25.7	30.6	31.1	28.2

2.2. Generation of samples from pilot-scale gasification

Details of the pilot-scale test program, mode of operation, and operating strategies can be found elsewhere along with supporting lab scale studies of the gasification behaviour of those coals [12,13]. The pilot-scale test program used a range of gasifier operating conditions that allow the performance of the coals to be compared under a consistent set of conditions and also allow the effects of changes in gasification conditions to be quantified for each coal in a comparable manner. Each coal was gasified under more than one set of steady state conditions, where samples of discharged quench water, process gas, and slag were taken. Solids present in the discharged quench water (referred to as process water solids, PWS) were also collected. Tapped slag was removed in batches from the lock-hopper system located at the bottom of the quench vessel. One slag sample from each test period (run) was analysed for its chemical and physical characteristics (including viscosity behaviour). If the carbon content of the sample was greater than 5 wt%, the carbon was removed from sample before conducting the analysis of ash composition and ash fusion behaviour by combustion at 800 °C. For SEM-EPMA analysis and for viscosity measurements, only coarse tapped slag, indicated in Fig. 2, was used.

2.3. Laboratory techniques

2.3.1. Laboratory slag preparation

Laboratory-prepared slags were produced to assess coal slag viscosity behaviour using techniques described previously [2]. Briefly, a sample of each coal was ashed in a muffle furnace in air at 800 °C. The compositions of these ash samples were analysed in accordance with Australian Standard Methods AS1038.15. In order to be consistent with the fluxed CRC703 used in the pilot-scale gasifier, the laboratory ash produced from CRC703 was mixed with flux at a rate consistent with that used in the trials (30 kg limestone to 1000 kg coal, equivalent to ~30% flux by weight of ash). The mixture was then homogenised with ring mill and pelletised. Samples were then melted in molybdenum crucibles under a nitrogen atmosphere at 1550 °C for use in viscosity measurements. To ensure reducing conditions, the crucible was placed in a sacrificial graphite liner. The compositions of the ash samples used for laboratory slag preparation are listed in Table 2.

2.3.2. Slag characterisation and analysis

Slag viscosity measurements were made using a Haake high-temperature viscometer using a rotational bob technique described