Gasification of Victorian Lignite in a Laboratory Scale Fluidised Bed Gasifier.

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Abstract

A 200-mm diameter, laboratory-scale atmospheric-pressure fluidised-bed reactor was designed and constructed by the Cooperative Research Centre (CRC) for Clean Power from Lignite. The purpose of this facility is to obtain experimental data for the air/steam gasification of Australian lignite in order to validate the Centre's mathematical model of a bubbling fluidised bed gasifier. An air-dried mixture of low-ash Victorian lignite has been used in air-steam and air-only gasification tests. The product syngas composition demonstrated successful gasification of coal with carbon monoxide and hydrogen concentrations each in the range 16-20 vol%. More carbon monoxide was measured in the syngas during coal gasification with air only. The gas composition of major species was observed to be relatively constant within the freeboard of the gasifier.

1 Introduction

The environmental, human and cost issues associated with energy use today require society to use resources, including fossil fuels, more efficiently. The large lignite deposits of Australia are a vital energy resource for this country, however the high inherent moisture content of these coals result in comparatively high energy-specific emissions of carbon dioxide during conventional power generation [1].

The Cooperative Research Centre (CRC) for Clean Power from Lignite is focusing on developing new power generation technologies using lignite, to increase generation efficiency and to reduce greenhouse gas emissions. Fluidised bed gasification is a key component in many of the new technologies being considered, whether for the re-powering of existing plants or for new advanced-cycle power generation plants.

In the design of these new technologies modelling tools are essential. A mathematical model of a bubbling fluidised bed gasifier has been previously developed by researchers within the Centre [2,3]. The model assumes a two-phase representation of the fluidised bed and considers the *net-flow* of gas from the emulsion phase to the bubble phase, which is important to correctly account for the substantial increase in molar gas flows that occur during the gasification of low-rank coal. The model has been validated, to some extent, against the limited data that is available in the literature. However it has not yet been rigorously applied to the gasification of Australian lignite.

Thus, a laboratory-scale fluidised bed gasifier has been designed and constructed [4] in order to obtain relevant and detailed experimental data for the gasification of Australian lignite and to validate the fluidised bed gasifier model against this data. The gasifier, described in detail below, has a 200

mm ID. reaction section with an expanded freeboard. It operates continuously and the design incorporates an interchangeable mode of operation between bubbling- and spouted-bed gasification, although only the bubbling-bed configuration has been tested to date. The intended experimental program is to investigate gasification of Victorian and South Australian lignite with air and steam, and with air only, as the gasification agents.

This paper reports preliminary results from the commissioning of the new experimental facility and from the first phase of gasification experiments using Victorian lignite.

2 Experimental methodology

2.1 Plant description

The fluidised bed gasifier is intended for air-blown and air-steam gasification of low rank coals at atmospheric pressure. The gasifier is designed to operate at temperatures up to 1000°C, with air flows in the range 100 to 350LPM (STP), steam flows up to 5.0 kg/hr and with coal feed rates less than 20 kg/hr. The main reaction section of the gasifier is 200 mm ID x 1.2 m high, extending into a 320 mm ID x 1.25 m high freeboard section. The freeboard section is covered with a spring-loaded, heavy steel cover which acts to provide pressure relief for the gasifier. A schematic diagram of the plant is presented in Fig. 1.

To compensate for heat losses, and to raise the temperature of the reactor during start-up, external electrical heating is provided in the form of 4x4kW cable type heating elements wrapped outside the gasifier, each with individual PID control. Air is supplied from the air compressor, metered through a mass flow controller and preheated by a Leister air tool to a maximum of 650°C. The air stream, if necessary, can be split before being heated to provide air injection to the freeboard. Steam is generated in a 6 bar electrical boiler and is separately metered to the gasifier plenum chamber. A drilled holed distributor plate is used to distribute air and steam across the

An initial batch of char is introduced to the bed via the batch hopper inlet for start up. Coal or char is then continuously fed to the reactor via a controllable reversible water-cooled jacketed screw feeder during gasification. The screw feeder and hopper arrangement is suspended via a winch and pulley arrangement and can be raised or lowered to relocate the feeder to one of several feed entry points. Pre-dried coal in 200L drums is loaded via a Hydrum drum lifter into the hopper auger and then conveyed to the coal hopper.

The gasifier is fitted with one primary cyclone followed by two smaller secondary cyclones for particulate removal. The gases then pass through two parallel water bubblers to cool

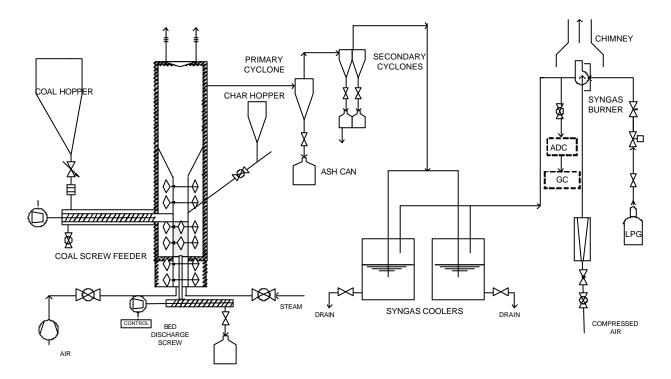


Figure 1: Schematic diagram of the 200-mm bubbling fluidised-bed gasifier.

and further remove particulates. The cooled syngas passes to the syngas gas burner, which is supported by a LPG pilot flame with a temperature control alarm system to detect flameout.

The gasifier plant is serviced by a closed loop circulating cooling water system with an air-cooled convective heat exchanger. Cooling water is used to cool the gas sampling probes and the coal feeder.

Discharge of solids from the gasifier is through a motor-driven speed-controlled water-cooled discharge screw, which removes bed material through a discharge pipe located in the centre of the air distributor.

Gasification experiments are commenced by first heating the gasifier to approx. 650°C using preheated air and the external heating elements. The gas flow is then switched to nitrogen and a batch of coal char is loaded to the gasifier from the char hopper via a designated chute. Once the char reaches approximately 600°C, air is reintroduced to the reactor at a controlled rate and continuous coal feeding from the coal hopper is initiated. After reaching the desired operating temperature, steam flow is introduced to the reactor.

The operation and monitoring of the gasification process involves controlling air and steam flows, monitoring temperatures inside the reactor, gas composition, pressures across reactor and the sustainability of the syngas flame. Data for the process pressures, flows, temperatures and heating element currents are automatically recorded.

2.2 Gas analysis

A key feature of the gasifier design is the ability to sample gases not only from the freeboard, but also directly from the bed. Syngas is drawn under vacuum from a sample location after the syngas coolers and prior to the syngas burner. The sample passes through an in-line particle and condensate trap and via an electrically heated sampling line to an ADC system, which consists of a heated particulate filter, perma-pure drier and infra-red CO and CO₂ analysers. The discharge from the ADC system is also fed to a Micro Gas-Chromatograph (GC), which measures species concentrations for nitrogen, oxygen, methane, hydrogen, hydrocarbons, sulphur species, carbon dioxide and carbon monoxide at 2-minute intervals.

Both, the ADC and Micro GC are used to continuously monitor the post-bubbler syngas concentration during a gasifier operation. Additionally, the Micro GC is used to analyse gas samples taken via water-cooled gas sample probes from the in-bed and freeboard gasifier zones using Teflon sample bags. Nine probes are available for use at any of 15 separate sampling locations spaced over the height of the reactor. The probes are blown clear using either compressed nitrogen or compressed air and allowed to be purged with syngas from the gasifier which operates at a slight positive pressure. Sample bags are then attached to each of the designated probes and are allowed to fill simultaneously.

3 Results and Discussion

Several air/steam and air blown only gasification tests of lignite coal were carried out in the 200 mm gasifier rig, mostly at 850°C. The coal used in these tests was an 80/20% mixture of Victorian Loy Yang and Yallourn coals, respectively and the coal was screened to between 1 and 3.35 mm to remove the majority of fine material. An analysis of the coal is presented in Table 1.

Of the twelve tests conducted, the most valuable data comes from the five most recent tests, since the earlier tests were undertaken during the commissioning of the gasifier. The results of these later tests are presented in Table 2. For Tests 1-7, the bed was loaded with a commercial char, Auschar, made from Yallourn coal, with a particle size of 0.5 - 2 mm.

In subsequent tests (Tests 7-12), the initial char bed was loaded with char of the Loy Yang/Yallourn coal, with a particle size of 0.25-3.35 mm. This char was collected as discharge from the gasifier during the preceding test, ie. the char remaining from one test was used as the initial char bed for the next test. The overall conversion efficiency (dry basis), determined from the total amount of dry coal fed to the gasifier as compared to amount of coal recovered in the bed material and from the cyclones at the end of each test, is also shown in Table 2.

Table 1: Ultimate analysis, and moisture content, of the Loy Yang /Yallourn coal mixture.

Ultimate Analysis	% db
Ash	1.1
°C	68.4
Н	4.6
N	0.6
S	0.29
0	25.0
Cl	0.05
Na	0.06
Moisture, %	14.0

Table 2: Key operating and performance data for ar-

Test	Aver.	Free-	Coal	Air/	Steam	Super-	Syng	as aft	er coc	olers:	Coal, on	Press.	Bed
No	Bed	board	Feed	/Fuel	/Fuel	ficial					dry basis	diff.	depth
	Temp	temp.	Rate	Ratio	Ratio	velo-	CO	H2	∞	CH4	overall	across	est.
						city					conver.	bed	
											effic.	mm	
	℃	℃	kg/hr	kg/kg	kg/kg	m/s	%	%	%	%	%	H₂O	cm
7.1	850	854	7.2	2.27	0.56	0.66	15.4	18.7	13.2	1.4	-	54	26
7.2	855	860	7.2	1.80	0.56	0.57	14.2	20.6	14.0	1.8	-	50	25
7.3	846	850	7.2	1.80	0.56	0.57	13.5	20.3	14.0	1.8	-	92	48
7.4	842	850	7.2	1.80	0.56	0.56	14.2	20.0	13.9	1.7	84.5	100	50
8.1	854	860	8.7	2.35	0.40	0.75	17.0	17.2	11.2	1.2	-	74	41
8.3	892	890	12.0	1.66	0.00	0.61	21.0	17.5	8.5	1.4	80.1	86	48
9	850	859	8.0	2.04	0.40	0.62	16.0	18.5	12.0	1.3	81.6	104	54
10	850	858	8.7	1.88	0.38	0.63	16.5	20.5	12.3	1.5	78.9	110	57
11	832	840	8.0	2.34	0.41	0.68	12.8	15.2	13.5	1.2	79.0	110	57
12.1	840	850	8.7	2.01	0.42	0.67	14.2	17.0	13.5	1.3	-	96	50
12.2	837	848	8.7	1.78	0.00	0.46	16.3	14.9	11.4	1.8	-	104	55
12.3	837	848	8.7	2.18	0.44	0.72	13.6	18.4	14.6	1.5	83.4	145	75

blown, steam gasification of an 80/20 Loy Yang and Yallourn coal mixture in a fluidised bed.

3.1 Freeboard gas composition

Gas samples were collected from the freeboard at 3 locations: immediately above the bed, at $0.5\,\mathrm{m}$ above the bed and at 1 m above the bed. Figure 2 shows that for the conditions given for Test 7.4 and for a bed height of approx. 50 cm, the gas composition remains essentially constant throughout the freeboard. Similar results were observed for other test conditions with different Air/Fuel (A/F) ratios and different bed heights. The carbon monoxide (CO) and hydrogen (H₂) concentrations in the freeboard were measured in the range of 13-16 vol% for CO and 18-20 vol% for H₂.

However, Fig. 2 also shows that the gas composition in the freeboard differs from that measured within the fluidised bed. It is observed that the CO concentration in the bed is higher than that measured in the freeboard while the $\rm H_2$ concentration measured in the bed is lower than the freeboard concentration. These results suggest that the gas-phase water-gas shift

reaction is occurring to some extent prior to the gases in the bubble phase entering the freeboard.

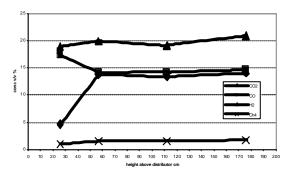


Figure 2: Syngas composition in the bed and freeboard during the air/steam gasification of 80/20 Loy Yang/Yallourn coal mixture. Bed height = 50 cm; Air/Fuel ratio = 1.8; Test 7.4.

3.2 Air/steam v. air only gasification

Gasification in air, with a lower Air/Fuel ratio compared with that used for air/steam gasification, was carried out as test 8.3 and 12.2. Significantly higher, up to 21 vol%(db), CO concentrations were observed during air gasification. Whereas the hydrogen concentration in the syngas during air-only gasification was either lower, in the case of Test 12, or approximately equal, in the case of Test 8, than that observed during air/steam gasification. These results can be explained in terms of the lower A/F ratio for the air-only gasification conditions and the lack of steam resulting in a reduction of hydrogen formation via the water-gas shift reaction. The bed temperature for Test 8.3 was significantly higher than for other tests and this resulted in the relatively higher concentration of CO.

3.3 Gas composition within the fluid bed

To examine variations of gas composition within the fluidised bed, for given gasifier conditions, up to nine gas samples were simultaneously withdrawn directly from the fluidised bed via water-cooled sample probes. Our experience in using this sampling method is that it is reliable and gives consistent results. However sampling probes located near the top of the bed, in the so-called "splashing" zone of the bed, were easily blocked and this would often hinder, or in some cases prevent, filling of the bag.

The results obtained by sampling directly from the bed clearly showed inter-bed variations in the gas composition. With increasing height above the distributor, there is a steady increase in both CO and H_2 , reaching a maximum near to the top of the bed. However the measured concentration of CO_2 was observed to initially drop, reach a minimum at some location within the bed, and then to rise again to levels similar to those measured immediately above the distributor. Similar variations in the inter-bed CO_2 concentrations have been observed, to some extent, in all tests. For one test, radial variations in concentration within the bed were examined via a traverse of the water-cooled sample probes. The radial gas concentrations were observed to be uniform and thus the extent of radial protrusion of the sample probes was determined to be insignificant.

In Test 12 the influence of bed height on the in-bed gas composition and on the syngas composition was examined. The bed temperature during the test was constant at approximately 850°C, with the bed height initially maintained at approximately 50cm and then increased to 75cm. (Note that bed height is estimated based on ?P across a small section of the bed.) The results showed similar trends to those in earlier tests in that a steady rise of CO and H₂ was observed within the bed, while the CO₂ concentration decreased in the lower part of the bed and then rose again in the upper part.

In relation to bed height, the results were rather inconclusive. Concentrations of CO measured along the fluid bed reached higher values at lower bed height, while H_2 concentrations were slightly higher for higher bed height, as shown in Fig. 3. It was also observed that the CO reached its highest concentration near the top of the bed, and H_2 at the very top of the bed or at the start of the freeboard, regardless of the total bed height. Obviously the water-gas shift reaction plays a role in the relative concentrations of CO and H_2 , however there are clearly other contributing factors and this demonstrates the importance of a good mathematical model in understanding the complex interactions within a bubbling fluidised bed.

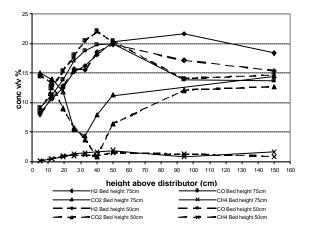


Figure 3: Syngas composition in the fluidised-bed during air/steam gasification of 80/20 Loy Yang/Yallourn coal mixture. Test 12.1: Bed height = 50 cm and A/F = 2.01 (dotted line); Test 12.3: Bed height = 75 cm and A/F = 2.18 (continuous line). Results plotted at 150cm correspond to composition after the syngas coolers .

In all tests we observe this characteristic 'dip' in CO_2 concentration with increasing bed height. The rapid formation of CO_2 , which occurs in the 5 cm of the bed located below the position of the first sample probe, is consistent with the fast consumption of O_2 due to combustion reactions. The rapid consumption of O_2 within the bed is confirmed by the Micro O_3 since O_4 detected at the first sample probe (5 cm above distributor plate) is always less than O_3 vol%.

Between 5cm and 10cm, the bed can be characterised as well-mixed. The high partial pressure of CO_2 , encourages CO_2 reaction with carbon via the Boudouard reaction:

$$C + CO_2 \Leftrightarrow 2CO$$

Consequently, the CO₂ concentration diminishes with a corresponding increase in CO. At some point in the bed, however, due to bed density and superficial fluid velocity, the

bed 'percolates' less and tends to 'bubble' more, forming pockets of gases that expand as they move through the bed. At this point, the gas phase equilibrium

$$CO + H_2O \Leftrightarrow CO_2 + H_2$$

shifts to the right and continues to do so, until the gases are ejected into the freeboard and equilibrium is attained in the splashing zone just above the bed.

3.3 Ash deposition

Despite the very low ash content of the coal used in this work, dislodged ash deposits were found in the bed material throughout the testing program. Heavily sintered deposits of up to 1 cm thick were observed to form on the distributor plate. Only an estimated 6% of the total ash input to the gasifier was found in these deposits.

4 Conclusions

Experience gained during the commissioning of a laboratory-scale continuous fluidised bed gasifier has resulted in steady reliable operation. Syngas composition demonstrates successful air/steam gasification of a Victorian low-rank coal with up to 21 vol% CO and 21 vol% H₂. Satisfactory results were also obtained using air-only gasification.

Analysis of gas samples withdrawn from within the fluidised bed and from the gasifier freeboard showed:

- A steady rise in CO and H₂ with bed height
- A rapid formation of CO₂ due to combustion reactions near to the distributor, followed by a gradual decrease in CO₂ to a minimum before increasing again in the top of the bed
- The gas composition in the freeboard is relatively constant
- Small radial variations in gas composition within the fluid bed

Future work will investigate steam/air gasification of high-sulphur, high-ash South Australian lignite.

5 Acknowledgments

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6 References

- Ashman, P.J. and Mullinger, P.J. Research issues in combustion and gasification of lignite, *Fuel*, 84, pp. 1195-1205, 2005.
- Yan, H-M., Heidenreich, C. and Zhang, D-K. Mathematical modelling of a bubbling fluidised-bed coal gasifier and the significance of 'net flow', *Fuel*, 77, pp. 1067-1079, 1998.
- 3. Ross, D.P., Yan, H.M., Zhong, Z. and Zhang, D.K. A non-isothermal model of a bubbling fluidised-bed coal gasifier, *Fuel*, In Press.
- 4. Ross, D.P. Fluidised bed gasification at Thebarton laboratory, *Proceedings of the 12th Annual Conference, CRC for Clean Power from Lignite*, pp. 185-189, June 2003.