Fluidized bed gasification of selected South African coals

by A.D. Engelbrecht*, R.C. Everson†, H.W.P.J. Neomagus†, and B.C. North‡

Transaction Paper

Synopsis

An investigation was undertaken to ascertain the suitability of four selected low grade, South African coals for gasification in a bubbling fluidized bed for production of synthesis gas and for the development of integrated gasification combined cycles (IGCC). This study consisted of the characterization of the coals, laboratory evaluation of intrinsic reactivity, and experimentation with a pilot-plant fluidized bed gasifier. Results of the characterization experiments show that the selected coals are high in ash, rich in inertinites, very dense (low porosity), with low caking indices and high ash fusion temperatures. Reactivity measurements with a thermogravimetric analyser (TGA), under reaction rate controlling conditions with carbon dioxide, showed that the reactivity of the coal chars decreases with increase in rank of the coal, as expressed by the vitrinite random reflectance and carbon content of the parent coals. The fixed carbon conversion achieved in the fluidized bed gasifier also correlates well with the rank parameter of the coal, with higher conversions being obtained with the lower rank coals. Thermal shattering and attrition of the coal particles produce significant amounts of fines, which correlate with the grindability indices, and no agglomeration (non-caking) was observed. It is concluded that fluidized bed gasifiers are able to utilize typical low grade, high ash South African coals for synthesis gas production and for inclusion in integrated gasification combined cycles for power generation.

Introduction

Coal is the most important energy source in South Africa, supplying approximately 75% of its primary energy. Due to the small reserves of oil and gas and the high cost of renewable energy such as hydro, wind and solar, coal will remain South Africa's most important energy resource for the foreseeable future. Based on scientific analysis it is generally accepted that a link exists between climate change and the use of fossil fuels such as coal. The development of clean coal technology (CCT) has therefore received increased attention worldwide with integrated gasification combined cycle technology (IGCC) being identified as a potential CCT that can be applied for power generation in the 21st century. With IGCC, power station efficiencies can be improved from the current 35% to between 45% and 55% and the emission of carbon dioxide, sulphur dioxide and nitrogen oxides to the atmosphere can be reduced more effectively (*in situ*).

In an IGCC power station, gasification replaces combustion as the primary coal conversion process. Fluidized bed gasification is a potential gasification process that can be utilized for IGCC plants in South Africa. The main advantages of fluidized bed gasification are that high-ash coals can be gasified, fine coal (< 5mm) can be utilized and no by-products (tar and oil) are produced. A disadvantage of the fluidized bed gasifier is that due to the lower operating temperature the carbon conversion is lower than entrained flow and fixed bed (Lurgi) gasifiers, which operate at a higher temperature. The lower carbon conversion can decrease the IGCC power station efficiency. Another disadvantage of fluidized bed gasifiers is that attrition, agglomeration and clinkering of coal particles can result in defluidization of the bed, which requires shutdown of the gasifier to allow clinker removal from the furnace.

Four high-ash South African coals were therefore selected to determine how coal properties under suitable operating conditions, affect fluidized bed gasifier performance in terms of carbon conversion, gas composition (calorific value), thermal cracking and attrition, and bed stability with respect to agglomeration/clinkering. This paper accordingly presents[1] characterization results of the different coals, which also include relative intrinsic reactivity results,[2] description, and performance results of a pilot-scale fluidized bed gasifier using very similar operating conditions for all the experiments.

* *Council for Scientific and Industrial Research, Pretoria.*

† *School of Chemical and Minerals Engineering, North-West University, Potchefstroom Campus.*

‡ *CSIR.*

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Experimental

Origin of coal samples

The four coals selected for the study were those that would be likely feedstocks to IGCC power stations in South Africa in future. The criteria used for selection of the coals for the study are that they should be high in ash (30–45%), feed coals to existing Eskom power stations, and the estimated lifetime of the mines producing the coals should be between 30 and 50 years. New Vaal, Matla, Grootegeluk and Duvha coals satisfied the above criteria and were accordingly selected for this study. Geographical, technical and historical information on the selected coals is given in Table I.

Characterization of coal samples

The characterization of the coal samples consisted of: proximate and ultimate analysis, calorific values, vitrinite reflectance, maceral analysis, structural analysis (BET adsorption), grindability indices (Hardgrove), caking properties, ash melting temperatures, chemical analysis of residual ashes, and the relative intrinsic reactivity of the coal samples. The methods used for the determination of the chemical and physical properties are well documented and the reference codes are indicated in Table III. The analyses were carried out by Advanced Coal Technologies (Pretoria), Petrographics SA (Pretoria), South African Bureau of Standards (SABS Pretoria) and Protechnik Laboratories (Pretoria). The BET (Brunauer, Emmert and Teller) analysis involved the adsorption of nitrogen. A very rapid TGA-based (thermogravimetry) method was used to determine the relative intrinsic reactivity of the coal samples[1,2]. The gas flow rate through the TGA was adjusted to ensure the absence of film diffusion and the overall reaction was controlled by the gas-solid reaction rate by using a pure carbon dioxide (slow reaction)[3]. This determination was considered as an initial rapid scan of the relative gasification potential (ranking) of the coals, exclusively for the chemical reaction rate (and structure). The TGA used was a Bergbau-Forschung GmbH7 model supplied by Deutsche Montan Technologie (DMT), Germany. A schematic representation of the apparatus is shown in Figure 1, which is described in detail by Everson *et al.*[3].

Fluidized bed gasifier

A pilot-scale fluidized bed gasifier (FBG) at the CSIR was used to investigate the gasification performance of the four selected coals in terms of carbon conversion and gas quality. A flow diagram of the FBG pilot plant is given in Figure 2 and specifications of the FBG pilot plant are given in Table II.

Coal, air and steam are the input streams to the process, which produce the output streams of gas and char (ash). Coal is fed to the gasifier by means of a screw conveyor at a height of 1.5 m above the distributor. Steam is generated in an

Table I

Information on the selected South African coals

	New Vaal	Matla	Grootegeluk	Duvha
Location of mine	Free State Province	Mpumalanga Province	Limpopo Province	Mpumalanga Province
Coalfield	Sasolburg	Highveld	Waterberg	Witbank
Production rate (Mt/a)*	15.2	14.0	15.0	16.0
Started production	1985	1979	1985	1979
Expected lifetime (years)	30–40	40–50	40–50	30–40
Coal preparation	Washed	Raw coal	Washed	Raw coal
Receiving power station	Lethabo	Matla	Matimba	Duvha
Power station rating (GWe)	3.6	3.6	3.6	3.5

*2005

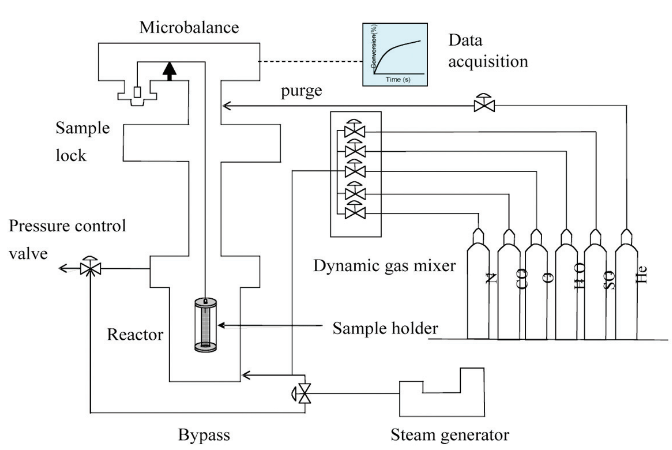

Figure 1—Schematic representation of the themogravimetric analyser apparatus

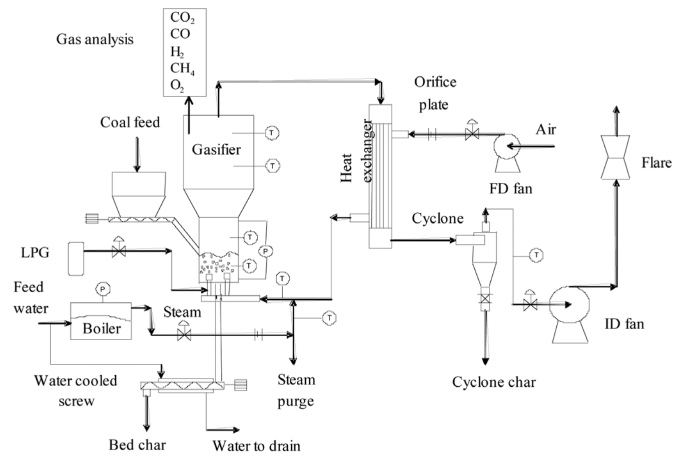

Figure 2—Flow diagram of the fluidized bed gasifier pilot plant

Table II

Specifications of the FBG pilot plant

Operating pressure	Atmospheric
Bed dimensions (m)	0.2 × 0.2 (square)
Freeboard dimensions (m)	0.55 × 0.55 (square)
Furnace height (m)	4 (2 m bed & 2 m freeboard)
Fluidized bed height (m)	< 0.6
Coal federate (kg/h)	18–30
Coal particle size (mm) (d_{50})	1–2.5
Coal CV (MJ/kg)	> 10
Air flow rate (Nm^3/h)	40–60
Steam flow rate (kg/h)	5–12
Bed temperature (°C)	850–950
Air temperature (°C)	155–210
Fluidizing velocity (m/s)	1.5–2.5

electrode boiler and is mixed with preheated air before introduction into the gasifier via the distributor. The gas produced during the gasification process is used to preheat the air, using a shell-and-tube heat exchanger. Char is removed from the bed (bed char) by means of a water cooled screw conveyor and from the gas (cyclone char) by means of a cyclone which is placed after the gas cooler. The de-dusted gas is combusted (flared) before it is vented to atmosphere.

The FBG is started up by using a silica sand bed which is heated by means of an LPG burner. When the bed temperature reaches 650°C, coal addition to the furnace is started. When the temperature reaches 850°C, LPG injection is stopped, the pilot flame (lance) is removed, and the furnace door is closed. The furnace is operated in combustion mode for 6 h in order to heat the refractories and freeboard. After 6 h, the coal flow rate is increased ± fourfold and steam is added to produce reducing conditions (oxygen deficient) in the furnace. The furnace is operated for a further 6 h to allow the bed carbon content and freeboard temperature to stabilize. Once stable conditions have been achieved, operating data are recorded and samples are collected for a period of 3 to 4 h.

Results and discussion

Chemical and physical properties

The results are shown in Table III. The coals are bituminous according to a ranking procedure based on vitrinite reflectance, and classified as a low grade coal (Grade D) with high ash contents and low calorific values. The New Vaal coal has the lowest rank parameter (vitrinite reflectance) being closer to the sub-bituminous coals and the Duvha coal has the highest rank parameter being closer to the semi-anthracite coals. The high inertinite content, which is characteristic of South African coal reserves, indicates that dense chars (low porosity)[5] will be formed during gasification with a corresponding low overall reactivity as a result of restricted intraparticle penetration of reacting gases.

The surface area and porosity from the BET adsorption measurements are very low, which is a result of the presence of a large fraction of very dense minerals (ash) together with the naturally occurring macerals.

The Hardgrove grindability indices obtained, which is an indication of the resistance to fragmentation (grinding) of the coal particle, are all within the range over which fines will be produced [ISO 5074]. The free swelling index (FSI) and Roga index (RI) reported, which are parameters used to assess caking and agglomerating properties (tendency to deform and stick together) of coal indicate that only the Grootegeluk coal could cake and/or agglomerate. The high ash fusion temperatures obtained indicates that all of the coals are favourable for fluidized bed operation since no melting of the ashes and the forming of clinkers would occur.

Intrinsic reactivity characterization

Reactivity determinations of the coals with a Thermo-gravimetric analyser were carried out at temperatures similar to that used in the fluidized bed gasifier (875°C to 950°C) and at atmospheric pressure (87.5 kPa) with a reaction gas consisting of pure carbon dioxide (100%). The results obtained from the TGA consisted of a short pyrolysis period followed by a long char gasification period and finally a period characterized by constant weight corresponding to the mass of the residual ash. The gasification period was considered in the study for the assessment of relative reactivities and was calculated from the thermogrammes which displayed distinct transition breaks between the periods. Also the results were expressed as carbon conversion versus time on an ash-free basis[2]. Typical char results for all the coal samples at 95°C are shown in

Table III

Chemical and physical characterization of coal samples

Property	Method	New Vaal	Matla	Grootegeluk	Duvha
Calorific value					
Calorific value (MJ/kg)	ISO 1928	15.1	18.6	19.8	21.0
Proximate analysis					
Inherent moisture (%)	SABS 925	5.80	3.50	1.60	1.80
Ash content (%)	ISO 1171	40.4	33.4	34.9	32.5
Volatile matter (%)	ISO 562	19.20	21.00	24.90	19.90
Fixed carbon (%)	By diff.	34.60	42.10	38.60	45.80
Ultimate analysis					
Carbon (%)	ISO 12902	42.58	50.66	51.96	58.70
Hydrogen (%)	ISO 12902	2.19	2.65	3.15	3.33
Nitrogen (%)	ISO 12902	0.89	1.07	0.99	1.27
Sulphur (%)	ISO 19759	0.69	0.74	1.58	1.10
Oxygen (%)	By diff.	7.54	7.97	5.85	3.14
Maceral content (daf) and vitrinite reflectance (rank)					
Liptinite (%)	ISO 7404-3	3	5	7	4
Vitrinite (%)	ISO 7404-3	23	38	52	13
Inertinite content (%)	ISO 7404-3	74	57	41	83
Vitrinite reflectance (%)	ISO 7404-3	0.53	0.64	0.68	0.75
Structural properties					
BET surface area (m^2/g)	Nitrogen adsorption	7.01	2.08	<1	< 1
BET Porosity (%) pore diameters < 810 Å	Nitrogen adsorption	3.2	1.3	1.5	1.1
Other physical properties					
Hardgrove grindability index (HGI)	ISO 5074	66	51	47	60
Free swelling index (FSI)	ISO 501	0	0	1	0
Roga index (RI)	ISO 335	0	0	10	0
Ash fusion temperature (°C)	ASTM D 27	1600	1500	1500	1600
Bulk density (kg/m^3)	Displacement	1700	1620	1570	1577

Table IV

Reactivity indices (h^{-1}) of chars

Char	Temperature (°C)				Rank parameters of parent coal	
	875	900	925	950	Vitrinite random reflectance (%)	C (%) dry ash free
New Vaal	2.75	3.80	5.63	8.92	0.53	79.1
Matla	0.51	0.65	0.99	1.56	0.64	80.2
Grootegeluk	0.14	0.27	0.40	0.75	0.68	81.8
Duvha	0.13	0.21	0.34	0.52	0.75	89.4

Figure 3. The results show clearly that the reactivities over the complete range of conversions are very different and that it correlates with the vitrinite reflectance and carbon content. (Table IV), which is in agreement with many published results[4]. This dependence can also be seen in terms of a reactivity index defined as $R_s=0.5/t_{0.5}$ with $t_{0.5}$ the time (hours) for the char to reach a conversion of 0.5[5,6] as shown in Table IV for all temperatures examined.

Pilot-scale fluidized bed gasification

Pilot scale gasification tests were carried out on the four selected coals using the bubbling fluidized bed gasifier (FBG) described above. All experiments were done at ± 925°C and ± 950°C (mid-bed) and at atmospheric pressure. The results of eight fluidized bed gasification experiments consisting of plant measurements, gas analysis and calculated variables are shown in Table V. The fluidizing velocity was more than three times the calculated minimum fluidizing velocity and low enough to prevent excessive carry-over of unconverted char to the cyclone. The gas calorific value was calculated from the composition and heats of combustion of the components of the gas. The fixed carbon conversion and char elutriated were calculated from carbon and ash balances respectively and the residence time reported (average value) was determined by assuming a perfectly mixed fluidized bed.

All experiments were carried out at approximately the same char residence times, bed temperatures (mid-bed), particle sizes and fluidizing velocity, since the objective of the

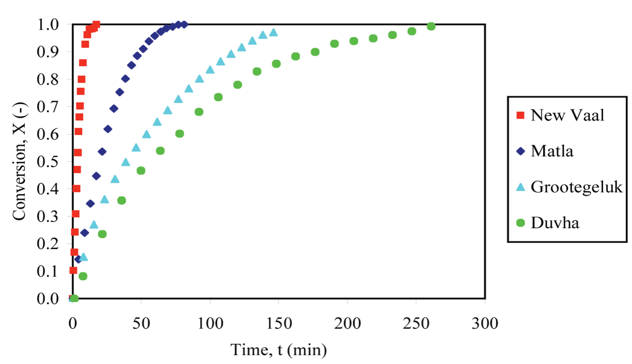

Figure 3—Char reactivities at 950°C from thermogaravimetric analyser

Table V

Fluidized bed gasification experiments at 90 kPa absolute pressure

	New Vaal		Matla		Grootegeluk		Duvha	
Experiment number	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**
Coal feedrate (kg/h)	28.7	23.9	27.0	24.3	23.0	23.0	26.4	26.4
Airflow (Nm^3/h)	52.2	47.0	50.6	50.9	48.5	47.8	47.5	47.8
Steam flow (kg/h)	9.1	5.8	8.5	8.5	10.2	10.0	10.9	9.0
Air and steam temp. (ºC)	202	159	190	185	173	178	176	186
Oxygen: carbon molar ratio	0.48	0.52	0.42	0.47	0.46	0.45	0.34	0.35
Steam: carbon molar ratio	0.50	0.38	0.41	0.46	0.57	0.56	0.47	0.39
Coal particle size (mm)[1]	2.4	1.2	1.6	1.6	1.9	1.9	1.6	1.6
Fluidizing velocity (m/s)[5]	2.2	1.9	2.1	2.2	2.1	2.1	2.1	2.1
Mid-bed temperature (ºC)	922	947	925	949	927	953	927	949
Lower bed temperature (ºC)	967	948	995	972	948	978	921	954
FBG exit temperature (ºC)	750	720	752	756	742	764	761	773
Dry gas composition								
CO (%)	NR[2]	11.1	10.8	11.6	8.7	10.2	8.8	9.9
H_2 (%)	NR	8.6	10.0	9.6	9.4	9.5	8.5	9.3
CH_4 (%)	NR	0.7	0.8	0.7	1.1	1.1	0.8	0.7
CO_2 (%)	NR	15.8	14.8	14.6	15.0	14.9	15.3	15.0
N_2 + others[3] (%)[4]	NR	63.7	63.5	63.4	65.7	64.2	66.5	65
O_2 (%)	NR	0.1	0.1	0.1	0.1	0.1	0.1	0.1
CO/ H_2	-	1.29	1.08	1.21	0.92	1.07	1.03	1.06
Gas calorific value (MJ/Nm^3)[5]	-	2.8	3.0	3.0	2.7	2.9	2.5	2.7
Bed pressure drop (Pa)	2664	2115	2553	2259	2553	2553	2455	2456
Char residence time (min)[5]	36.7	36.6	37.4	37.6	45.1	45.1	35.7	35.7
Carbon in bed char (%)	2.8	1.4	24.3	20.8	26.8	26.4	38.6	33.9
Bed char particle size (mm)	1.1	0.6	1.2	1.0	0.8	1.0	0.9	1.2
Carbon in cyclone char (%)	19.5	15.5	32.3	27.8	31.0	27.0	41.6	43.2
Cycl. char particle size (mm)	0.07	0.08	0.05	0.07	0.07	0.07	0.06	0.08
Char elutriated to cyclone (%)[5]	60.6	66.6	53.8	55.6	51.1	51.6	58.3	59.9
Fixed carbon conversion (%)[5]	82.7	85.9	68.2	74.0	63.2	67.0	52.0	53.7

[1]d_{50}—50% of the coal mass is less than the d_{50} size
[2]NR—no reading
[3]Others are < 0.4 % and include H_2S, NH_3, HCN and C^{2+}
[4](N_2 + others) by difference
[5]Calculated results.

investigation was to evaluate the performance of four different coal feedstocks. With the start-up procedure described above and the conditions given in Table V the fluidized bed was found to attain a steady state condition 6 h after start-up during which measurements were made. Evidence of this is given in Figure 4 where the gas concentrations are plotted against time in Figure 4.

The fixed carbon conversion varies significantly for the four coals between 52% and 85.9% and was also found to correlate with the vitrinite reflectance and carbon content, as shown in Figure 5. This trend was also obtained with the results from the thermogravimetric experiments with very different reaction indices (reactivity), which is attributed to the different modes of operations, such as differential (low conversion), isothermal conversion with reaction controlling in the thermogravimetric analyser as opposed to non-isothermal, bubble and emulsion phase operation with coupled diffusion effects, and particle attrition in the fluidized bed. A similar result was obtained by Jing *et al.*[7] who gasified three bituminous coals of differing rank originating from Chinese coalfields in a fluidized bed gasifier. It should be noted that the carbon in the fines has a higher carbon content than the bigger particles in the bed. Also, by increasing the temperature from 925°C to 950°C the fixed carbon conversion increases by an average of 3.6%.

The gas compositions and consequently the gas calorific values for the different coals depend only slightly on the coal type at the operating conditions, and it was not possible to identify any trends. Similar results have been published by Gururanjan and Argarval[8].

Thermal shattering and attrition of the coal particles in the bed also occurred with the generation of smaller particles (average diameter) in the bed and fines that are eventually elutriated from the gasifier. The relative size reduction can be seen in Table V. The relationship between the percentage cyclone char and the Hardgrove grindability index (HGI) is given in Figure 6 for each coal and temperature. The char

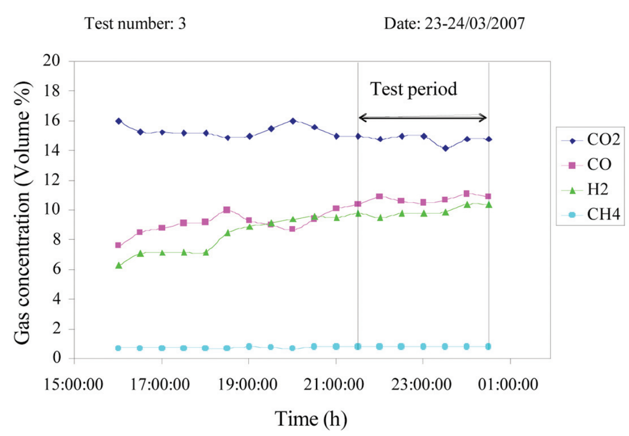

Figure 4—Gas concentration versus time for Malta coal at 925°C

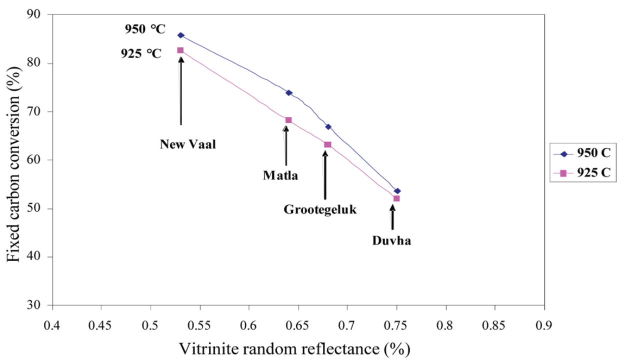

Figure 5—Fluidized bed gasifier fixed carbon conversion versus reflectance (rank)

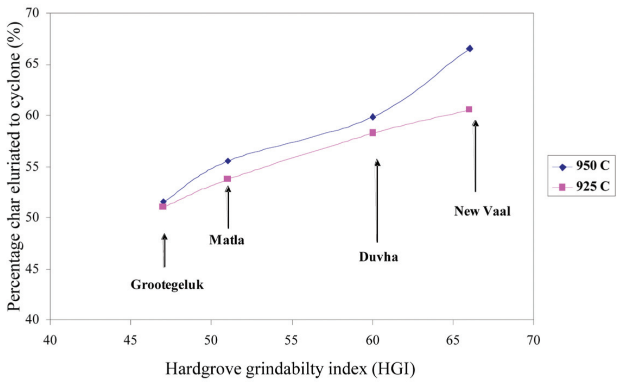

Figure 6—Char fines generated as a function of Hardgrove grindability index

fines generated and elutriated increase with the grindability of the coal and with the temperature. Using the experimental results and least squares regression, a correlation was developed to predict the amount of cyclone char that would be produced by the gasifier. This is given by the following equation:

$$\textit{Elutriated char}\ (\%) = 3.13(HGI)^{0.69}\ (U)^{0.33}\ (d_p)^{-0.19}$$

with dp = the mean particle size (d_{50}) of the feed coal, U = the fluidizing velocity and *HGI* = the Hardgrove grindability index of the coal. A similar correlation was used by Rhinehardt *et al.*[9] to predict the carry-over of fines.

From Table V it can be seen that the temperature close to the distributor (T1) is higher than the temperature in the middle of the bed (T3). This is a result of oxygen reacting with the char and with back-mixed combustible gas close to the air distributor[10]. Ciesielczyk and Gawdzik[11] reported that the maximum temperature is limited to the bottom 10% of the bed and is up to 200°C higher than the uniform bed temperature in the middle of the bed. The maximum temperature increase observed during the pilot plant experimentation was ± 70°C (experiment 3).

Due to the non-caking nature of the coals tested (Table III), bed agglomeration and defluidization of the bed did not occur during any of the experiments. The same result was achieved by Gutierez and Watkinson[12] with Canadian coals in a fluidized bed gasifier.

Conclusions

A pilot-plant fluidized bed gasifier (coal feed rate 18–30 kg/h) was designed and was successfully operated with selected low grade coals with carbon conversion ranging from 54% to 86% at 950°C for approximately the same set of operating conditions[13]. The results from the laboratory experiments using a micro-reactor (intrinsic reactivity) and results from the pilot plant (overall reactivity) all correlate with coal rank (vitrinite reflectance and carbon content) of the parent coal, with the coal with lowest rank having the highest reactivity. This indicates that the characteristics of the coal and resultant chars have a significant affect (controlling) on the overall performance of the fluidized bed in addition to other transport and mechanisms characteristic of fluidization. Published results in the literature on the dependence of coal/char reactivity on rank have been deduced mostly from experimentation with ideal laboratory reactors[7]. The amount of fines generated in the gasifier and the resulting elutriation rate of fly ash from the gasifier can be described well by the Hardgrove grindability index (HGI) of the feed coal. The low free swelling (FSI) and Roga (RI) indices, together with the high ash fusion temperature (AFT) of the coals tested, prevented agglomeration, clinkering and defluidization of the bed. Due to the relatively low gasification reactivity of most South African bituminous coals, a secondary char combustion stage may be required after the fluidized bed gasifier (at optimal conditions), in order to achieve overall carbon conversions in excess of 95%.

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