FLUIDISED BED GASIFICATION OF LOW GRADE SOUTH AFRICAN COALS CSIR/MSM/EP/EXP/2006/0101/A

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ABSTRACT

To cope with increasing electricity demand, South Africa will need to install approximately 1000 MW of generating capacity per year for the foreseeable future. Whilst there is increasing pressure to adopt non-fossil fuel electricity generating technologies, it is a fact that South Africa will continue to exploit its abundant coal reserves. The challenge, therefore, is to utilise the coal as efficiently as possible, in order to reduce carbon dioxide emissions.

This paper describes an on-going investigation into one potential Clean Coal Technology (CCT), namely fluidised bed gasification. Coal gasification holds the potential benefits of increased efficiency, reduced water consumption and co-production of liquid and gaseous fuels and chemicals.

A suite of five South African coals has been identified as being possible fuels for power stations which would operate for three or four decades, towards the middle of this century. These coals are being subjected to thermogravimetric analysis (TGA) to ascertain their reactivity under gasification conditions. Further, pilot scale gasification trials will be carried out on two of these coals. The objective is to rank the coals in terms of suitability for gasification, and to ascertain if TGA analysis can give an accurate prediction of actual performance in a gasifier.

Keywords: coal, gasification, fluidised bed, reactivity, characterisation

1 Introduction

Coal is the most important energy resource in South Africa since 71% of our primary energy and 88% of our electricity is derived from coal. 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 our most important energy resource for at least the next 75 years

Coal gasification is regarded as the most likely technology to replace conventional coal combustion for power generation in the 21^{st} century. With coal gasification, power station efficiencies can be improved from 35% currently to between 45% and 55%. The emissions of CO₂ into the atmosphere can also be reduced by capture and sequestration.

A potential disadvantage of coal gasification compared to coal combustion is that the rate of coal conversion is slower due to the reducing condition in the gasifier. This can result in low coal conversion efficiencies particularly when low reactivity coals are used. The low coal conversion efficiency results in a net efficiency drop for an Integrated Gasification Combined Cycle (IGCC) power station.

It is therefore important to characterise South African coals in terms of reactivity under gasification conditions so that suitable coals can be identified and the right gasification technology can be matched to a specific coal.

2 Integrated Gasification Combined Cycle Technology

The flowsheets for conventional and IGCC power generation cycles are given in Figure 1. In a conventional cycle all the energy in the coal is used to generate steam which is then exhausted through a steam turbine to generate electricity. The exhaust steam has to be recondensed and recycled to the boiler. Due to large condensation losses the overall efficiency (coal to electrical power) of a conventional power station is between 33% and 38%. This can be raised to 40%-45% by increasing the temperature and pressure of the steam. New high performance steels need to be developed to achieve this target.

In an IGCC power station a coal gasifier is incorporated in the flowsheet. During gasification coal is reacted with oxygen/air and steam to produce a combustible gas (syngas). This gas stream is relatively easy to clean since it is under pressure and has a low volume compared to flue gas resulting from conventional coal combustion. The cleaned gas is combusted in a gas turbine that produces electrical power while heat is recovered from the turbine exhaust gas by means of a conventional steam cycle. This configuration (IGCC) produces higher efficiencies 45% - 55% and lower emissions than conventional power stations.



The energy flows in an IGGC power station is given in Figure 2.

Figure 1: Conventional and IGCC power generation.



Figure 2: Energy flows in aIGCC power station.

3 Hybrid Gasification - Combustion Systems

Due to the low carbon conversions (70% - 85%) that are achieved in fluidised bed gasifiers, hybrid combined cycle systems are being developed which combine features of both coal gasification and combustion.



Figure 3: Hybrid gasification combustion system.

From Figure 3 it can be seen that uncombusted char and fly-ash from the gasifier is fed to the combustor with additional coal to convert the residual char. The partial gasifier and combustor are pressurised fluidised beds.

The advantages of this system are:

- The overall carbon efficiency of the system is greater than 99% without the need for long residence times in the gasifier
- Cleaning of the hot combustible gas in a ceramic bag filter increases the efficiency of the gas turbine and therefore the overall coal to electrical power efficiency of the cycle
- The flue gas is desulphurised by means of limestone feeding into the gasifier
- Steam is not required in the partial gasifier since flue gas from the combustor is injected for temperature control.

Due to the relatively low reactivity of South African coal the hybrid gasification combustion system could be a potentially attractive clean coal technology (CCT) option for South Africa to pursue in the future.

4 Coal gasification

Coal gasification is a key enabling technology for IGCC systems [1]. Coal gasification is not new to South Africa since Sasol operates 72 Lurgi gasifiers (since 1981) at its synfuel plants in Secunda. The Lurgi gasifier however uses coarse coal (-50 mm + 12 mm) and is more suited to synthetic fuel and chemicals production. For IGCC plants fine coal (< 6 mm) gasification is the technology of choice [1]

	Fluidised bed	Entrained flow
Coal particle size	0.5 mm – 5 mm	0 – 0.5 mm
Coal moisture	Dry	Dry/slurry
Coal type	Non-caking coals	Any coal
Ash in coal	< 60%	< 30%
Gasification agents	Air/steam/oxygen	Steam/oxygen
Gasification temperature	850°C – 950°C	1300℃ – 145 0℃
Pressure	0 - 10 bar	0 - 30 bar
Residence time	0.5 – 1.5 hrs	< 10 s
Carbon efficiency	70% - 85%	75% - 90%
Gasification efficiency	60% - 75%	55% - 65%
Commercial examples	HTW,KRW, U -gas	Texaco, Prenflo, Shell

 Table 1: Comparison of fluidised bed and entrained flow fine coal gasifiers.



5 Properties of South African power station coal

The objective of our project is to assess the suitability of South African coal for fluidised bed coal gasifiers. Five coals were selected that are feeds to existing power stations in South Africa. The proximate analysis of these selected coals is given in Table 2.

	Calorific	Ash	Moisturo	Volatila	Fixed	Total	Sizo
	Calornic	7.511	MOISture	Volatile	i ixeu	Total	0126
	value	content		matter	carbon	sulphur	grading
	(MJ/kg)	(%)	(%)	(%)	(%)	(%)	(mm)
New Vaal	15.56	40.2	5.9	22.3	31.6	0.55	0 - 6
Grootegeluk	20.66	33.8	2.5	26.7	37.0	1.48	0 - 5
Matla	20.48	27.1	5.3	24.1	43.5	1.08	0 - 8
Syferfontein	19.69	28.3	5.6	22.0	44.1	0.75	0 - 6
Duvha	21.85	30.3	2.3	20.6	46.8	0.97	0 - 6

Table 2: Proximate analysis of typical power station feed coal in South Africa.

It can be seen that these coals have high ash contents and low calorific values and are therefore low grade.

Other coal properties important for fluidised bed gasification are:

- Coal reactivity in atmospheres of CO₂ and H₂O
- Caking index and free swelling index (FSI)
- Ash fusion temperature (AFT)

5.1 Coal reactivity

The gasification reactions (1 and 2) in a gasifier occur at a much lower rate (up to 1000 times slower) than the combustion reaction (3)

$C + CO_2$	\rightarrow 2CO	(1)
$C + H_2O$	$\rightarrow \text{CO} + \text{H}_2$	(2)
C+ O ₂	$\rightarrow CO_2$	(3)

The reactivity of coal therefore has a major effect on the carbon conversion efficiency that can be achieved in a fluidised bed gasifier.

The reactivity of coal is affected by structural properties of the coal, which include the surface area and porosity, and the intrinsic reactivity is dependent on the surface chemistry and the catalytic effect of inorganic compounds. [3]

As the gasification reactions proceed the char pore structure changes continuously with extent of reaction which leads to variations in the effective area for reaction and, then, to variations in reactivity. [6]

The reactivity of coals can be measured by means of a thermogravimetric analyzer (TGA) using CO₂ or steam as the reacting gas.

A Mettler TGA/SDTA 851e at the University of Pretoria and a Bergbau – Forshung TGA at the NWU was used to measure the reactivity of the coals given in Table 2.

During a TGA experiment the weight loss of the char is measured as a function of time for a fixed temperature and CO_2 concentration.

The conversion (X) as a function of time (t) and conversion rate (dX/dt) as a function of conversion (X) are presented in Figures 6 and 7 for the five selected coals.



CHAR CONVERSION VS TIME

Figure 6: Char conversion as a function of time.

The relative reactivity of coal (relative to other coals) is often compared by using the reactivity index R_s [4, 5] which can be expressed as:

$$R_{s} = \frac{0.5}{\tau_{0.5}}$$
(4)

with $\tau_{0.5}$ being the time (hrs) for the char to reach a fractional conversion of 0.5. Therefore if the char takes 30 minutes to reach a fractional conversion of 0.5 the reactivity index is 1. The reactivity index of the coals tested is given in Table 3 below.

The rate of char conversion is often expressed using the rate equation given below. [6, 7]

$$\frac{dX}{dt} = k_0 \exp(\frac{-E}{RT}) P_{CO_2}{}^{\alpha} (1 - X)^{\beta}$$
(5)

 k_0 - Pre exponential factor (min⁻¹)

- *E* Arrhenius activation energy (J/mol)
- R Universal gas constant = 8.314 (J/mol.K)
- P_{CO_2} Partial pressure of CO₂ (kPa)
- lpha Reaction order with respect to the gas
- β Reaction order with respect to the solid

CHAR CONVERSION RATE VS TIME



Figure 7: Rate of char conversion as a function of conversion.

For the above tests the temperature and CO_2 partial pressure was held constant. Equation 5 therefore reduces to:

$$\frac{dX}{dt} = k (1 - X)^{\beta}$$
(6)

k - Rate constant (1/min)

The values of R_s , k and β for the five coals tested are given in Table 3.

	R _s (hrs⁻¹)	k (1/min)	β
New Vaal	3.02	0.067	0.87
Syferfontein	1.82	0.038	0.52
Matla	1.68	0.037	0.44
Grootegeluk	1.51	0.031	0.81+
Duvha	0.92	0.012	1.29

Table 3: Reaction parameters for coals tested.

+ For X < 0.92 * For X < 0.78

The two most popular models to describe the way gas reacts with solid particles is the shrinking core model and the homogenous model (also referred to as the volumetric model)

[4]. The shrinking core model assumes that the reaction occurs at the external surface of the particle and gradually moves inside leaving an ash layer behind. The homogenous model assumes that the reaction takes place uniformly throughout the whole volume of the particle. The actual reaction normally takes place via both of the above models simultaneously. A lower value of β indicates that the shrinking core model is the dominant mechanism [4]. Coal particle size will have a greater effect on the reaction rate if the particle reacts via the shrinking core model.

The reactivity index and reaction rate constants obtained (Table 3) are typical of high ash inertinite rich coals.

The reactivity index of two Chinese bituminous coals [6] measured under similar conditions as reported in Table 3 gave reactivity indexes of between 5 and 6 indicating that these coals are more reactive than the coals tested.

The effect of CO_2 pressure (Bergbau – Forshung TGA at the NWU) on reaction rate for two of the selected coals is given in Figures 8 and 9.

From Figures 8 and 9 it can be seen that the conversion rate of char increases with partial pressure according to equation 5. For constant temperature the equation becomes:

$$\frac{dX}{dt} = kP_{CO_2}{}^{\alpha} \left(1 - X\right)^{\beta} \tag{7}$$

	DX/dt	k	α	β
	(1/min)⁺	(1/min.bar)		
New Vaal 1 bar	0.0187	0.034895	1.000	0.9
New Vaal 5 bar	0.0278	0.034895	0.175	0.7
Grootegeluk 1bar	0.0036	0.009036	1.000	1.36
Grootegeluk 5 bar	0.0075	0.009036	0.365	1.08
+ Y-05		•		

Table 4: Values of k, α and β in equation 7.

X = 0.5

From Table 4 it can be see that the char conversion reaction is first order with respect to CO_2 partial pressure up to 1 bar. Above one bar the reaction order with respect to CO_2 tends to zero.

CHAR CONVERSION VS TIME



Figure 8: Char conversion vs time at 1 and 5 bar pressure.



CHAR CONVERSION RATE VS TIME

Figure 9: Char conversion rate vs conversion at 1 and 5 bar pressure.

This is consistent with findings from other investigators [9] who have studied the effect of pressure on gasification reactions. Above 15 bar no further increase in the rate is observed with increasing pressure.

5.2 Caking index

The caking index of coal refers to the tendency of the coal chars to become soft when heated to high temperatures (> 600°C). If the coal chars become soft they tend to stick to surfaces and to one another. The free swelling index (FSI) is closely related to the caking index i.e. coals that tend to swell on heating also become soft and sticky.

Coals with a high caking index can cause poor mixing and agglomeration in fluidised bed gasifiers resulting in eventual de-fluidisation of the bed. For fluidised bed gasification of low grade South African coal it is therefore important to characterise South African coal in terms of caking index and FSI.

6 Pilot Plant Testing

Pilot plant tests are required to correlate the reactivity parameters of the coals tested with the carbon conversion efficiencies that can be achieved in an actual fluidised bed gasifier. Pilot plant testing is also required to relate the caking index of the coals to agglomeration and sintering in the gasifier.

A flow diagram and specifications of the CSIR pilot fluidised bed gasifier is given in Figure 10 and Table 5.



Figure 10: Flow diagram of the CSIR Pilot fluidised bed gasifier.

Bed dimensions (m)	0.2 × 0.2
Bed area (m ²)	0.04
Fluidised bed height (m)	< 0.5
Freeboard dimensions (m)	0.5 × 0.5
Freeboard area (m ²)	0.25
Furnace height (m)	4 (2m bed & 2m freeboard)
Coal feedrate (kg/h)	20 to 30
Coal particle size (mm)	< 5
Coal CV (MJ/kg)	15 to 25
Air flowrate (Nm ³ /h)	25 to 45
Steam flowrate (kg/h)	5 to 10
Bed temperature (°C)	850 to 1000
Air temperature (°C)	100 to 200
Fluidising velocity (m/s)	1 to 2.5
Gas CV (MJ/Nm ³)	3 to 4
Pressure	Atmospheric
Operating mode	Combustion & Gasification
Gas cleaning	Cyclone

Table	5:	Specificati	on of the	CSIR	pilot	fluidised	bed	gasifier.
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Table 6: Fluidised bed pilot plant results

Coal	Coal flow (kg/h)	Air flow (Nm ³ /h)	Steam flow (kg/h)	Bed Temp. (℃)	Carbon in ash (%)	Carbon Efficiency (%)
New Vaal	22	37	0	920	14	83
Syferfontein						
Matala		I	In progress			
Grootegeluk						
Duvha						

7 Discussion and conclussions

Thermogravimetric analyser (TGA) tests conducted on five South African power station coals show that:

- The coals have a relatively reactivity in the range of 1 to 3
- The reactivity is lower than overseas coals that are of a low grade.

Pilot plant test are required to:

- Ascertain if TGA tests can give a good prediction of actual performance in a gasifier
- Ascertain if the caking index and free swelling index (FSI) can be used to predict agglomeration, de-fluidisation and clinkering of the fluidised bed.

7 References

- 1) Izzo L., "Calpine fuels diversity initiative", Calpine fuels corporation communication.
- 2) Yang Y., and Watkinson A.P., "Gasification reactivity of some Western Canadian coals", Fuel 73, (1994), 1786 -1791.
- Everson R.C., Neomagus H.W.J.P., Kasaini H., and Njapha D., "Reaction kinetics of pulverized coal-chars derived from inertinite-rich coal discards: Gasifcation with carbon dioxide and steam", Fuel 85 (2006), 1076 -1082.
- Zang L., Huang J., Fang Y., and Wang Y., "Gasification reactivity and kinetics of typical Chinese anthracite chars with steam and CO₂", Energy and Fuels 20, (2006), 1201 -1210.
- Ye D.P., Agnew J.B., and Zhang D.K., "Gasification of South Australian low-rank coal with carbon dioxide and steam: kinetics and reactivity studies", Fuel 77, (1998), 1209 – 1219.
- 6) Chunnhua L., Tomokazu W., Makoto N., Shigeyuji U., and Toskinori K, "Gasification kinetics of coal chars carbonized under rapid and slow heating conditions at elevated temperature", "Journal of Energy Resource Technology 123, March 2001, 21 -26.
- 7) De Carvalho R.J., and Brimacombe J.K., "Relationship between reactivity and surface area changes during the Boudouard reaction for low-rank Western Canadian coals".
- 8) Riley R.K., Judd M.R., and Wright D.W., "Steam gasification kinetics of Bosjesspruit coal char".
- 9) Sha X.Z., Chen Y.G., Cao J., Yang Y.M., Ren D.Q., "Effects of operating pressure on coal gasification", Fuel 69, (1990), 656 659.