GAS COMPOSITION AND TEMPERATURE WITHIN A SPOUTED BED GASIFIER

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ABSTRACT

Internal gas composition and temperature profiles have been measured in a spouted bed coal gasifier. These data are essential to develop a mathematical model that can be used for scale-up of the system. As a first step towards developing a kinetic model, the treatment of Yoshida and Kunii for the fluidized bed gasifier has been adapted to the spouted bed using the one-dimensional and the streamtube gas flow models.

Two Western Canadian bituminous and sub-bituminous coals having particle size -3.36 + 1.19 mm were gasified to produce low and medium calorific value gas. The effect of different operating variables including total bed height, average bed temperature and char recycling on the internal gas composition profiles were investigated. In a typical run eighteen gas samples were collected from different levels and radial positions in the bed. In addition the average axial temperature profiles along different sections were also recorded.

Both radial and axial gas composition profiles exist in a spouted bed coal gasifier. The combustion reaction takes place in a very narrowzone close to the gas inlet in the annulus and in the lower section of the spout. The temperature variation in the spout was large, and the upper section of the spout was found to be the hottest region in the bed. A hydrogen/carbon monoxide ratio of about unity has been observed throughout the bed for the system producing low calorific value gas. In this case the contribution from pyrolysis and volatile reforming reactions on the gas composition profiles appears to be significant, especially in the upper section of the bed. Gasifying char instead of coal altered the carbon monoxide and hydrogen distribution in the bed. Less hydrogen was produced. This pattern was however reversed when oxygen rather than air was used to gasify coal and was partly because of the high steam/oxygen ratio.

Both the one-dimensional and the streamtube models gave relatively similar predictions of the average gas composition profiles and the overall gas composition based on kinetic expressions from the literature. However the predictions could not be matched with the experimental results. In modelling, the particle movement in the bed should be described. Pyrolysis should not be assumed instantaneous but its kinetics should be included. The streamtube model was superior to the one-dimensional model since it was able to predict radial composition profiles throughout the annulus. These profiles however were much steeper than those found in the experiments.

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1. INTRODUCTION

The direct use of solid fuel is limited and its use is less attractive as compared to gaseous and liquid fuels in many applications. The reserves of solid fuel, especially coal, are abundant and extensive research has been directed towards developing suitable processes that can efficiently utilize this material. An example of such a process which is still being developed is gasification.

Coal gasification involves the production of gaseous fuels such as methane, hydrogen and carbon monoxide by allowing coal to react with steam and/or carbon dioxide at elevated temperatures. As in any other process involving reactions of solids and gas, high conversion of coal in gasification depends mainly on the gas-solid contacting technique used. A number of reliable gasifiers based on various gas-solid contacting techniques have been developed and some of these have been commercially proven. These include moving, entrained and fluidized bed gasifiers. However there exist some disadvantages for different types of gasifiers which result in superiority of one with respect to the other depending on a number of factors such as the type of coal to be processed, particle size to be employed etc. Both moving and fluidized bed gasifiers suffer from a common disadvantage which is the inability to handle caking coal. Entrained bed gasifiers can accommodate caking coal, however they can only be operated using fine particles, which brings with it an economic penalty because of the cost of pulverizing Due to these problems, the spouted bed gasifier is being the coal.

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investigated with the hope of providing a stable and reliable technique for processing caking coals which are too coarse for entrained bed gasifiers.

The overall performance and the sensitivity to different operating variables have been studied for spouted bed gasifiers (13-16). Promising results have been obtained when compared to other units. However no information is available concerning the gas composition within the gasifier. To provide a scale-up based on principles of reactor design, some knowledge of the composition within different zones of the spouted bed is required. Since this information must be generated experimentally, an in-bed study of the gas composition profile is required. Previous work on hydrodynamics clearly indicated that the performance of the spouted bed as a chemical reactor is strongly influenced by the bed diameter. As relatively large bed diameters would be employed for commercial spouted bed gasifiers i.e. diameters much larger than those being investigated currently, development of a mathematical model is essential for scale-up purposes. A reliable model can only be developed using information obtained from an experimental study of the internal composition profiles in a spouted bed coal gasifier.

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2. LITERATURE REVIEW AND BACKGROUND

2.1 The Spouted Bed as a Chemical Reactor

The spouted bed has been developed as an alternative fluid-solid contacting technique to the fluidized bed and has recently received much attention for industrial applications due to its capability of handling relatively coarse particles. A complete review on this subject is given by Mathur and Epstein (1) and will not be presented here. In brief the spouted bed consists of a centrally located dilute phase concurrent-upward transport region (spout) surrounded by a dense-phase moving packed bed with countercurrent fluid percolation (annulus), as shown schematically in Figure 1.

A number of research papers on the application of the spouted bed as a chemical reactor have been published including the development of appropriate mathematical models for the system. The models generally treat gas phase reactions which are catalysed by solids. In some cases experimental results were used to verify the models and to identify the validity of the various assumptions used.

Mathur and Lim (2) introduced a one-dimensional model, where plug flow of gas was assumed both in the spout and the annular regions which were treated as two separate regions. The effect of gas back-mixing was neglected. The effect of the different operating variables on the overall conversion in a first order reaction was examined theoretically based on this approach. It was found that the overall extent of conversion of gas improved as the column diameter was increased and this

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Figure 1 - Schematic diagram of a spouted bed.

improvement was mainly due to an increase in conversion in the annulus. In all cases the extent of conversion in the spout was not significant. The role of spout voidage in determining the overall conversion was relatively unimportant even for the fastest reaction. Measurements made by Lim (3) showed that the spout voidage ranged typically from unity to 0.65 at z = H. Poorer results were obtained by increasing the operating gas velocity above U_{ms} . An improved overall conversion was obtained by increasing the bed height up to a certain value. Above this optimum height no significant changes were observed for slow reaction while for fast reaction, the conversion actually became poorer.

Based on the observation made in a half-sectional column with nitrogen dioxide used as tracer, Lim and Mathur (4) proposed a more realistic streamtube model as a modification to the one-dimensional two region model. In this model plug flow of gas with dispersion along curved streamlines in the annulus was proposed, as supported by residence time distribution measurements. Assuming the validity of this assumption, one would expect to observe a radial concentration profile in the annulus of a spouted bed chemical reactor. Vertical plug flow of gas in the spout, as assumed in the one-dimensional model, was also assumed in this case.

In an attempt to verify the two models proposed by Mathur and Lim (2,4), Piccinini et al (5) studied the decomposition of ozone on an iron catalyst in a 0.15 m diameter column. Unlike Mathur and Lim (2) where all the hydrodynamic features were predicted from equations, these investigators used experimentally measured values of all the requisite

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quantities except U_a and ε_s as input to the models. In addition, the original one-dimensional model was modified by allowing diffusional mass transfer between the spout and the annulus in addition to transfer by bulk flow. The overall conversion predicted by both models was in good agreement with the experimental values, with an average deviation of about 5%. The inclusion of a non-zero mass transfer coefficient between the spout and the annulus in the one-dimensional model gave poorer results.

Littman et al (6) examined the one-dimensional model proposed by Mathur and Lim (2) and theoretically simulated a first order reaction using the bed hydrodynamic equations different from those of the original study. The findings reported by Mathur and Lim were confirmed except for the effect of bed height on overall conversion where no optimum value was observed.

Further work on assessing the performance of spouted bed as a chemical reactor was performed by Rovero et al (7), where the decomposition of ozone on an iron oxide catalyst was examined in 0.15 m and 0.22 m diameter beds. To aid in discrimination between the two proposed models both the radial and longitudinal profiles of ozone concentration were determined. As in all previous cases the overall conversion predicted by both models was in good agreement with experimental values. However the one-dimensional model failed to account for the radial concentration profiles observed while the streamtube model predicted a much steeper concentration gradient over most of the annulus. Contrary to expectations and findings reported by

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all previous investigators, the effect of increasing bed diameter was to decrease the conversion. The results for the other variables investigated were in accord with the predictions made by Mathur and Lim (2) and by Littman et al (6).

Even though the two models suggested by Mathur and Lim (2,4) were able to give good overall predictions for most cases investigated, they failed to describe closely the annulus concentration profiles observed. As suggested by Rovero et al (7), the failure may be due to the assumption of no radial dispersion which could be significant especially in large columns. In addition, depending on the size of column, the conical bottom region may form a large fraction of the bed where there is evidence of circulation (8). Rovero et al (9) have shown that the superficial annulus gas velocity in the conical region is in excess of that predicted by the Mamuro-Hattori flow distribution (1) which has been used in most work.

It seems therefore that any realistic model for in-bed composition must account for radial concentration gradients in the gas phase.

2.2 The Coal Gasification Process

Previous work on coal gasification producing either low, medium or high BTU fuel gas employed a variety of different reactors including moving, fluidized and entrained beds. Each of these reactor types has its own characteristics which will influence the operating conditions to be selected and the type of coal to be used. Thus there exist

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advantages/disadvantages of one type of reactor over the other depending on the required operating conditions (10-12). The counter current direct contact heat exchange between gas and solids that takes place within a moving bed gasifier results in a relatively high overall conversion. However fine particles (< 3 mm) and caking coals cannot be handled by this type of reactor. The entrained bed reactor on the other hand can accept any type of coal and gives high throughput especially when operated at elevated pressure. Unfortunately the coal particle size must be smaller than about 75 µm and carbon conversion may be low. The fluidized bed gasifier has a relatively uniform bed temperature due to its excellent mixing characteristics. However, a disadvantage of this reactor is that difficulties are usually encountered when caking coals are used. This is due to the agglomeration that takes place, forming larger particles which cause the bed to collapse.

The spouted bed coal gasifier was developed with the purpose of providing a smooth and a reliable system for gasifying caking coal. The problems of agglomeration could be overcome with the high gas velocity in the spout which tends to break up any agglomerates that might form within the bed, and the absence of a grid where agglomerates can build up.

Systematic investigation of spouted bed coal gasification was initiated by group of investigators at the University of British Columbia. Foong et al (13) studied the effect of various operating parameters on the quality of gas produced in a 0.15 m diameter gasifier blasted with air, using inert bed of silica (1.2 - 3.4 mm). The various

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parameters investigated were bed temperature (1020 - 1220 K), coal feed rate (2.5 - 12 kg/h), steam feed rate (16 - 24 kg/h), particle size, bed height (0.25 - 0.6 m) and different types of coal (sub-bituminous Forestburg and highly caking Sukunka). Under the range investigated improved gas quality was obtained as the coal feed rate, bed temperature and the particle size were increased. Increasing the bed height up to a certain value improved the quality of gas produced but no further improvement was observed on increasing the height above this value. The coal feed rate was found to be an important factor in determining the smoothness and stability of the overall operation, especially when gasifying caking coal. In assessing the effect of coal feed location, these investigators concluded that bottom feeding of coal minimizes the presence of tar in the product stream as compared to feeding the coal at the top of the bed.

The fluid mechanical characteristics of spouted beds strongly depend on the column diameter. Hence a higher coal conversion and improved gas quality can be obtained when using a larger diameter gasifier. Watkinson et al (14-15) investigated the performance of a 0.3 m diameter gasifier. A number of operating parameters were examined in addition to those investigated previously. The effect of char recycle and particle size distribution were studied at coal feed rates of about 50 kg/h. The conclusions of the previous investigators were confirmed in this work. Char recycle showed no significant improvement in the gas quality produced, but the efficiency over once-through operation improved. The gasifier efficiency was reported to be significantly

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reduced by having a higher percentage of finer particles in the coal feed. This was mainly due to a large extent of particle elutriation from the bed. The advantage of the 0.3 m diameter gasifier over the smaller 0.15 m diameter was clearly demonstrated when a highly caking Sukunka coal was used. In this case a much higher percentage of carbon in the bed can be tolerated (\approx 37%) as compared to 10% for the smaller system. Other advantages such as higher gas calorific value, thermal efficiency and steam decomposition have been achieved from the bigger system. These gains were not solely related to the diameter increase itself.

Watkinson et al (16) compared the performance of the spouted bed to the already established fluidized bed. In their experimental work, the effect of air/coal ratio and coal feed rate, keeping the other parameters constant, on the gas heating value, total gas yield etc. were investigated under both modes of operations using different types of coal. They concluded that no appreciable differences between spouted and fludized bed existed as far as yield and heating value were concerned. These workers however suggested that the advantage of one over the other could exist if the systems were to be tested using other variables such as using oxygen rather than air, recycling of char and using the optimal particle size for each system. In addition, advantages or disadvantages may become obvious when comparisons were made using larger size reactors.

The performance of the spouted bed gasifier investigated was considered promising when compared to the other systems as shown in Table 1.

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Comparison of gasification processes*

»	Fluidized Bed		Martin D. I		
Process	Bamag-Winkler (17)	Westinghouse (18)	– Moving Bed Wellman-Galusha (17)	Spoul (13	ced Bed & 15)
Diameter (m)	5.5	0.5	3.05	0.15	0.305
Bed Depth (m)	3			0.40	1.0
Temperature (c)	800-1000	930-1020	900	800-930	805-860
Coal Throughput (kg/s•m ²)	0.136	0.32-0.60	0.123	0.188	0.23
Gaş Yield (m ³ /kg coal)	4.45		4.00	3.05	3.22
Gas Calorific Value (MJ/m ³)	3.91	3.7-4.5	5.97	3.6	4.36
Coal Type	Sub. bit A.	Sub. bit to Eastern bit.	High Volatile bit A.	Sub b (Fores	lt stburg)

*Air and steam blown. All at atmospheric pressure except Westinghouse at 1545 KPa.

2.3 Theory of Coal Gasification

2.3.1 Pyrolysis

Upon heating, coal undergoes physical and chemical changes to an extent which depends on a number of factors such as heating rate, temperature, coal type etc.

Pyrolysis or devolatization is normally the first step through which such major changes take place. During this process there is a substantial weight loss of coal due to the evolution of volatile matter which cannot be described by a simple single step reaction but occurs in stages involving a series of complex reactions, producing both tar and gases (CO_2 , CO_2 , CH_4 , H_2 and other liquid hydrocarbons). Depending on the environmental conditions, these products can undergo further secondary reactions involving cracking, polymerisation and deposition onto the char. The proportions and distribution of gaseous components and liquid yield is generally determined by both heating rates and the temperature zone of pyrolysis which can be classified into different categories as given in Table 2. Typically, under flash pyrolysis condition with minimum vapour residence time a large proportion of tar and unsaturated hydrocarbon will be produced (19). A number of investigators (20) have reported that volatile yields significantly greater than those indicated by proximate analysis can be obtained from coal by flash pyrolysis. In general, at any particular heating rate, both the total yield and gas/tar ratio increases with temperature. At a relatively low final temperature ($\simeq 500$ °C) increasing the heating rate will increase the total volatile yield while the gas/tar ratio

Table 2

Classification of heating rates and temperature zone of pyrolysis (19).

A. Classification of Heating Rates

		Heating Rate °C/s	Heat-Up Time to 1000°C for ~ 100 µ in Size
1.	Slow heating	<< 1	20 min
2.	Intermediate heating	5 ~ 100	10 s ~ 4 min
3.	Rapid heating	500 ~ 100,000	10 ms ~ 2 s
4.	Flash heating	> 10 ⁶	< 1 ms

B. Classification of Temperature Zones for Carbonization of Coal

Low temperature carbonization ~ 500°C
 Intermediate temperature carbonization ~ 750°C
 High temperature carbonization ~ 1000°C
 Very high temperature carbonization > 1200°C

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decreases. On the other hand, rapid heating at a much higher temperature ($\simeq 1000$ °C) will increase both the total yield and gas/tar ratio. Several investigators (21-22) working on flash pyrolysis reported that the tar yield shows a maximum value at about 600 °C.

Pyrolysis is a complex process and a number of simplified kinetic models have been proposed to describe the reaction. According to Anthony et al (23) primary volatiles formed are of two categories: reactive and non-reactive. The non-reactive volatiles escape completely from the particle, while only a portion of the reactive volatiles The remainder deposits inside the coal particle by escape. polymerisation. Chang et al (20) working on flash pyrolysis reported that only a portion of the coal volatiles directly pyrolysed into gaseous product. The rest will escape as tar, which could subsequently crack into smaller gas molecules until the system is quenched. Gas cracking was considered not to be important. In interpreting his data, Wen and Chen (24) assumed that coal will initially devolatilise to give tar. A portion of this tar will escape, another portion will crack into gaseous components while the remainder deposits back onto the char particles. According to this model no gas will be evolved directly from coal by pyrolysis.

A simpler model of pyrolysis is normally more attractive for gasifier design purposes, as several reaction coefficients have to be determined for any one particular coal if the above described models are to be used. These coefficients are normally difficult to generate. A review on coal pyrolysis by Anthony and Howard (25) indicated that most

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investigators approximated the overall pyrolysis process as a first order decomposition with respect to the amount of volatiles V that still remain in the coal, i.e.

$$\frac{\mathrm{d}V}{\mathrm{d}t} = K' \left(V_{\infty} - V \right) \tag{2.1}$$

where K' = pyrolysis rate constant, S^{-1}

 $V_{\infty} = V$ as t $\rightarrow \infty$

The variation of K' with temperature is assumed to follow Arrhenius relationship expressed as follows:

$$K' = K'_{o} \exp(-E'/RT)$$
 (2.2)

Values of K'_{o} and E' are to be experimentally determined for each particular coal. The above equation however does not predict the product distribution of the volatile released. Solomon (26) used the form of equation 2.1 above to predict the amount of each component released. In this case V and V_{∞} represent the amount of the particular component at time t and t $\rightarrow \infty$ respectively. The rate parameters K'_{o} and E' will have different values for different components and coal. According to Howard (27) the sequence of volatiles being released (after drying) is: chemical water, carbon dioxide, carbon monoxide, higher hydrocarbons (tar, etc.), ethane, methane and finally hydrogen.

2.3.2 Char-Gas Reaction

The heterogeneous reactions between char and gases are the main reactions that take place in a gasifier. These reactions are normally slow when compared to the homogeneous gas-gas reactions, and their rates of reactions are of considerable interest and importance for gasification studies. The following set of linearly independent gas-solid reactions are normally considered in describing gasification processes:

Combustion

$$\psi C + O_2 + 2(\psi - 1) CO + (2 - \psi) CO_2$$
(2.3)

Gasification

$$C + H_2 0 \rightarrow C0 + H_2$$
 (2.4)

$$C + CO_2 + 2CO$$
 (2.5)

$$C + 2H_2 \rightarrow CH_4 \tag{2.6}$$

The char-oxygen reaction is the fastest among all the reactions considered. This reaction, which is exothermic, acts as the main heat source for the intermediate endothermic char-steam and char-carbon dioxide reactions. The char-hydrogen reaction is known to be the slowest of the above four gas-solid reactions that takes place in a gasifier. For comparison, the relative rates of all the four reactions considered are given in Table 3.

The equilibirum consideration of gasification reactions (2.4) - (2.6) has been the commonly used procedure to obtain an approximate

Table 🛛	3
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Approximate relative rates of the gas-carbon reactions at 800° and 0.1 atmospheric pressure (41)

Reaction	Relative Rates
$C - O_2$	1 x 10 ⁵
$C - H_2O$	3
$C - CO_2$	1
C - H ₂	3×10^{-3}

approachable conversion in a real system. The extent to which a given reaction will proceed towards the right under given conditions is reflected by the value of its thermodynamic equilibrium constant K. A large K value implies large equilibrium conversion; a small K implies little conversion to product. Variation of K values for graphite-steam, graphite-carbon dioxide and graphite-hydrogen reactions with temperature are given in Table 4. These K values are based on activities (K_a) and in this case the activity of graphite is taken to be unity. It is clearly shown from these tables that high temperature favours the gasification of char with steam and carbon dioxide. In contrast the char-hydrogen reaction is only favoured at low temperature and high pressure. The extent of this reaction is so small under a typical gasifying condition of 1 atm and 800°C and is normally neglected. This reaction has been ignored by other investigators (28-29) in their studies of coal gasification at atmospheric pressure.

The char-gas reactions can be classified into two categories: volumetric and surface reactions as discussed below:

2.3.2.1 Volumetric Reaction

Volumetric reaction is characterized by the chemical reaction as the rate controlling step in porous solids. Here the reacting gas diffuses into the interior of the particles causing the reaction zone to spread throughout the solid. As the reaction proceeds, an ash layer may build up at the outside surface of the particle whilst the reaction zone continues to shrink. Figure 2 shows typical carbon and gas concentration profiles as a function of carbon conversion.

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Table	4	

The variations of K_a values for the graphite-steam, graphite-carbon dioxide and graphite-hydrogen systems with temperature (19)

Temperature °K	K _{C-H2} O	K _{C-C02}	к _{с-н2}
298.16	1.005×10^{-16}	1.014×10^{-21}	7.902 x 10 ⁸
600	5.058×10^{-5}	1.870×10^{-6}	1.000×10^2
800	4.406×10^{-2}	1.090×10^{-2}	1.4107
1,000	2.617 x 10°	1.900 x 10°	0.0983
1,500	6.081×10^2	1.622×10^3	0.00256



Figure 2 - Concentration profiles of carbon and reacting gas as a function of carbon conversion for volumetric reaction. Many investigators (30-32) concluded that the internal surface area due to the inside pores of the particle plays an important role in volumetric reactions. Most of this internal area is attributed to the micropores which are in the size range of 10-50Å. This internal surface area varies as the reaction proceeds. It first increases to a maximum and then reduces to a value of zero at complete conversion as reported by Dutta and Wen (30). According to these workers each coal/char sample exhibits its own pore structure and the change of such structure with conversion and temperature gives rise to a characteristic rate-conversion curve for that individual coal or char.

The following rate expression has been formulated by Dutta and Wen (30):

$$\frac{\mathrm{d}x}{\mathrm{d}t} = \alpha_{\mathrm{v}} k_{\mathrm{v}} C_{\mathrm{g}}^{\mathrm{n}} (1 - \mathrm{x})$$
(2.7)

where

- x = fraction of carbon conversion of char due to char-gas reaction, -
- t = time, s
- α_{r} = relative pore surface area function, -
- $C_{a} = gas concentration, mol/cm³$
- $k_v = volumetric rate constant for char-gas reaction (cm³/mol)ⁿ·s⁻¹$ n = order of reaction

 α_v describes the change in pore structure and is a function of

conversion and temperature for each type of coal in question. Equation 2.7 is applicable when resistance due to gas diffusion inside the particle is negligible. At high temperature, this resistance may become appreciable and the concept of effectiveness factor n is introduced as given by the following expression for n = 1.

$$\eta = \frac{3}{M} \left(\frac{1}{\tanh M} - \frac{1}{M} \right)$$
 (2.8)

where

$$M_{.} = \phi_{0} [(1 - x) \alpha_{v}]^{1/2}$$

 $\phi_0 = r_0 \sqrt{k_v C_{so}/D_{eo}}$

 r_0 = outside radius of particles, cm

 C_{so} = carbon concentration of char at zero conversion, mol/cm³ D_{eo} = effective diffusivity in solid at zero conversion, cm²/s Thus the volumetric rate equation of a first order reaction is expressed, including the interparticle diffusion as

$$\frac{\mathrm{d}x}{\mathrm{d}t} = n\alpha_{v} k_{v} C_{g} (1 - x)$$
(2.9)

The reaction rate constant k_v is assumed to follow Arrhenius relationship (30) given by

$$k_{\rm w} = k_{\rm o} \exp(-E/RT) \tag{2.10}$$

2.3.2.2 Surface Reaction

This type of reaction normally takes place when the reacting gas can hardly penetrate into the interior of the solid particle, i.e. for impervious solids and/or when the overall reaction is gas diffusion controlled. For this type of reaction, the reaction zone is confined to the surface of the unreacted core which shrinks as the reaction proceeds, while the product solids or ash layer builds up on the outside. Typical carbon and gas concentration profiles for such a reaction is given in Figure 3.

The resistance to the reaction is made up of the combination of resistances due to mass transfer of gas in the boundary layer surrounding the particle, gas diffusion in the porous ash layer and the surface reaction itself. Levenspiel (33) has formulated this type of reaction known as the shrinking core model expressed as follows:

Rate =
$$\frac{dx}{dt} = \frac{3P_g}{M_w r_o \rho_c (\frac{1}{k_{diff}} + \frac{1}{k_{ash}} \frac{(Y-1)}{Y} + \frac{1}{k_c Y^2})}$$
 (2.11)

where

 M_w = molecular weight of carbon, g/mol. r_o = outside particle radius including ash layer, cm ρ_c = molar density of carbon in coal particle, mol/cm³ P_g = gas partial pressure, atm. k_{diff} = diffusional reaction rate constant, g/cm² atm s k_{ash} = ash film diffusion rate constant, g/cm² atm s k_s = surface reaction rate constant, g/cm² atm s


Gas concentration profile

Carbon concentration profile

Figure 3 - Concentration profiles of carbon and reacting gas as a function of carbon conversion for surface reaction. Y = r_c/r_o r_c = core radius at time t, cm

Depending on the system employed, the product or ash layer may segregate from the particles as the reaction proceeds. This is normally true for the spouted bed gasifier due to high attrition. In the absence of resistance due to the ash layer, the instantaneous reaction rate from Equation 2.11 reduces to

Rate =
$$\frac{3P_g}{\frac{M_w r_o \rho_c (\frac{1}{k_{diff}} + \frac{1}{k_s})}{\frac{M_w r_o \rho_c (\frac{1}{k_{diff}} + \frac{1}{k_s})}{s}}}$$
(2.12)

The mass transfer through the boundary layer is influenced by a number of factors, such as the flow condition of the bulk gas (i.e. laminar or turbulent) as well as the relative velocity between the gas and the particles. Turbulent flow will bring the oxidant to the vicinity of the particles surface while high relative motion between the gas and the solid will reduce effective thickness of the boundary layer and enhance transfer rate through it. The values of diffusional reaction rate constant, k_{diff}, can be calculated to a fair degree of accuracy using the following predictive equation (34-35):

$$k_{diff} = \frac{24(1 + 0.6 \text{ Re}^{1/2} \text{ Sc}^{1/3}) D_{AB}}{d_{p} R'T}$$
(2.13)

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where

Re = Reynolds number
Sc = Schmidt number
D_{AB} = diffusion coefficient of species A in B, cm²/s
d_p = particle diameter, cm
R' = universal gas cnstant (82.06 atm · cm³/mol .°K)
T = temperature, °K

The ash film diffusion rate constant depends on both the gas diffusivity and the voidage of the ash layer. According to Wen and Chaung (36) the value of k_{ash} can be roughly estimated by the following correlation:

$$k_{ash} = k_{diff} \epsilon^{2-3}$$
 (2.14)

where ε is the voidage of the ash layer.

The surface reaction rate coefficient is assumed to follow the Arrhenius law (37) expressed as

$$k_{s} = k_{so} \exp(-E/RT)$$
 (2.15)

values of k_{so} and E for each coal are to be determined experimentally.

In general, the shrinking core model does not fully represent the actual gas-solid reactions especially when dealing with coal/char particles which are normally porous. Investigation by Smith and Taylor (38) and Field (39) on char combustion and Dutta and Wen (30) on char-carbon dioxide reactions indicated that the reactions inside the particles do take place simultaneously with the external reaction. However the shrinking core model represented by Equation 2.11 is simple and practically useful for modelling purposes. A number of investigators (36, 40) employed this model in their work on coal gasification.

2.3.2.3 Char-Oxygen Reaction

Char-oxygen reaction is the fastest heterogenous gas-solid reaction that takes place in the gasifier (42). The reaction is exothermic and provides most of the heat necessary for the other endothermic gasification reaction and can be represented as

$$\psi C + O_2 \rightarrow 2(\psi - 1) CO + (2 - \psi) CO_2$$
 (2.16)

where ψ is a system constant which depends on the reaction conditions and determines the primary product distribution of CO and CO₂ in the combustion products. ψ takes a value of 2 when CO is the direct product of combustion and a value of 1 when CO₂ is the only product.

Several investigations to predict the combustion product distribution and hence the value of ψ as a function of temperature, particle size, carbon type and oxygen partial pressure (43-45) gave no quantitative conclusions. However there is a general agreement that both CO and CO₂ are the primary products of char-oxygen reaction, with smaller particle and/or higher temperatures favouring the formation of CO. Larger particles and/or lower temperatures favour the CO_2 formation. Arthur (46) investigated the combustion product distributions for two carbons of widely different reactivities (graphite and coal char) at atmospheric pressure and in the temperature range of 400°C - 900°C and found that Z, the ratio of $CO:CO_2$, is given by

$$Z = 2500 \exp(-6249/T)$$
(2.17)

In his experiment the oxidation of CO was suppressed using the inhibitor phosphoryl chloride. Using the value of Z given by Equation 2.17 Wen and Dutta (19) suggested a correlation to predict the value of ψ expressed as follows:

For $d_p \leq 0.005$ cm

$$\psi = \frac{2Z + 2}{Z + 2} \tag{2.18}$$

For 0.005 cm < $d_p \leq 0.1$ cm

$$\psi = \frac{1}{Z + 2} \left[(2Z + 2) - \frac{Z(d_p - 0.005)}{0.095} \right], \quad d_p \text{ in cm}$$

For $d_p > 0.1 \text{ cm}$
 $\psi = 1.0.$

There is a large volume of work published, dealing with the kinetic studies of char-oxygen reaction. Mulcahy and Smith (47) reported that the overall combustion rate for particles larger than 100 μ is diffusion controlled at temperatures above 1200°K. According to Smith and Tayler (38), coal/char particles of about 90 μ m in size

will burn under chemical reaction control rate up to about 750°K, whereas for smaller particles ($\approx 20 \mu$) the chemical reaction control regime extends up to a higher temperature of about 1600°K. Field (39) in his experiment using particle sizes 28 μ - 105 μ concluded that char burns both internally as well as externally. This conclusion was based on the observation that both particle density and mean diameter reduce as a function of char burn-off. This observation is generally expected when working with small particles as the chemical reaction controls the overall rate of reaction, thus allowing the reaction to proceed inside the particles. Large particles are normally employed in spouted bed (\approx 2 mm) where diffusion is significant. Under this condition, the assumption of surface reaction for char combustion is justified (42). The rate of surface reaction as given by Equation 2.12 can be used for this reaction. In this case P_g represents the oxygen partial pressure.

The order of reaction n, for char-oxygen reaction has been reported to have values ranging between zero and unity (42, 48-51). However, there is a general agreement based on a large volume of experimental data that the char combustion is first order with respect to oxygen partial pressure. Hence Equation 2.12 when applied to char combustion reduces to

Rate =
$$\frac{{}^{3P}_{0_2}}{{}^{M}_{w} r_{o} \rho_{c}(1/k_{diff} + 1/k_{s})}$$
(2.19)

The value of k_{diff} to be used in Equation 2.19 can be obtained with a fair degree of accuracy from Equation 2.13. The effect of

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relative velocity and hence the Reynolds number on k_{diff} is not very significant for particles in the pulverized fuel size range ($\simeq 100 \mu$) as concluded by Mulcahy and Smith (47). For such a condition Field et al (48) suggested a simpler expression to calculate the k_{diff} for char combustion given as follows:

$$k_{\text{diff}} = \frac{24\,\psi\text{D'}}{\text{R'T d}_{p}} \tag{2.20}$$

where R' = Universal gas constant $(82.06 \text{ atm} \cdot \text{cm}^3/\text{mol} \cdot \text{°K})$ D' = diffusion coefficient of oxygen $4.26 (T_{1800})^{1.75}/\text{P}, \text{ cm}^2/\text{s}$ P = total pressure, atm ψ = mechanism factor given by Equation 2.18 d_p = particle diameter, cm

Equation 2.20 can be applied without much error to the fluidized bed char combustion where the Re is normally small.

The rate of surface reaction for char combustion as a function of temperature has been determined involving char of different reactivity (37,38,48,52-53). Most of this work was conducted using particles in the pulverized coal size range. A number of these correlations suggested by different investigators are given in Table 5. According to Field (39), there is no variation in the rate of surface reaction with particle size. Field et al (48) reported approximate value of k_{so} and E/R for char-oxygen reaction which can be used together with Equation 2.15 to predict the rate of surface reaction for char combustion as a

Rate equations for carbon-oxygen reaction

Rate = $kP_{0_2}^n$

Ref	k	n	Size (µ)	Temp. Range °K	Particle Type	Remarks
37	1.34 exp (-16405/T _s) (g/cm ² .s)	0	22, 49, 89	630 - 1812	Brown coal	Experiments were performed in fixed and entrained bed reactors under oxygen partial pressure up to 0.2 atm. Rate given is based on total surface area and is the rate of surface reaction.
38	$-0.49 + (3.85 \times 10^{-4} T_s)$ (g/cm ² .s)	0	20 - 100	1400 - 2000	Low rank non swelling coal	The effect of mass transfer was eliminated by theoretical arguments. Rate equation is based on the external surface area of particle, and is the rate of surface reaction.
48	8710 exp (-17976/T _s) (g/cm ² •s)	1	varied	750 - 1650	varied	Effect of mass transfer was eliminated by using small particles and/or high gas velocity or correction made to the observed rate for this effect using theoretical relationship. Rate equation is based on the external surface area, and is the rate of surface reaction.

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Table 5 continued...

Ref	k .	n	Size (µm)	Temp. Range °K	Particle Type	Remarks
71	20.4 exp (-9596/T _s) (g/cm ² ·s·atm)	1	6, 22, 49, 78	1400 - 2200	semi anthracite	Experiment was performed in a laminar flow reactor using N_2 - 0_2 mixture with variable 0_2 concentration of $0.1 - 0.2$ atm. Diffusion effect was taken into account from theoretical relationship. Rate equation is based on the total surface area, and is the rate of surface reaction.
51	55 exp (-20200/T) (g/cm ² •s•atm) ^s	1	6, 22, 49, 78	1400 - 2200	semi anthracite	Data obtained from referece 71 was used to correlate rate equation based on external surface area, and is the rate of surface reaction.

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function of temperature. These are

$$k_{so} = 8710 \text{ g/cm}^2 \cdot \text{atm} \cdot \text{s}$$
$$E/R = 17,967^{\circ} \text{K}$$

2.3.2.4 Char-Carbon Dioxide Reaction

Based on a number of studies in this area, the rate of char-carbon dioxide reaction can be correlated using the Langmuir-type adsorption equation expressed as follows (54-56).

Rate =
$$\frac{k_1 P_1}{1 + k_2 P_2 + k_3 P_3}$$
 (2.21)

Both P_1 and P_3 are the partial pressures of carbon dioxide and P_2 is that of carbon monoxide. According to this equation carbon monoxide would have an inhibiting effect on the char-carbon dioxide reaction. This was found to be the case as reported by Wen and Lee (19) and Batcheleder et al (54). The constant k_1 depends on the rate of adsorption of carbon dioxide on carbon, k_2 is the equilibrium constant for the adsorption-desorption of carbon monoxide on the carbon surface and k_3 depends primarily on the rate of reaction of the adsorbed carbon dioxide molecules on carbon as well as on the value of k_1 . These rate constants are however difficult to generate from experimental data as they are interrelated and become less attractive for gasifier design.

Dutta and Wen (30) investigated the rate of this reaction for a

number of coal samples at different sizes (-20 + 35, -60 + 100, -35 + 60, -100 mesh) in the temperature range of 840 - 1100 °C. They concluded that the reaction is best described by volumetric reaction according to Equation 2.7. On the other hand, Amundson et al (40, 57-58) and Wen and Chaung (36) assumed the applicability of surface reaction for the char-carbon dioxide reaction in their work.

The order of reaction for the char-carbon dioxide reaction has been found to have values ranging from zero to unity with respect to the partial pressure of carbon dioxide depending on the operating temperature and pressure. Wen and Dutta (42) suggested that a first order reaction can be assumed at low pressure up to 1 atm. As the pressure is increased typically above 15 atm, zeroth order reaction is approached. The reported values for the activation energy lie in the range of 55 - 65 kcal/mol. A typical value of 59 kcal/mol has been reported by Dutta and Wen (30). Differences in the values published are mainly due to the different reactivity of coal used under different experimental conditions.

A number of rate equations for the char-carbon dioxide reaction correlated by different investigators are given in Table 6.

2.3.2.5 Char-Steam Reaction

The mechanism for the char-steam reactions has been found to be similar to the char-carbon dioxide reaction (54,59) and the rate equation can be correlated by the Langmuir adsorption isotherm, represented by Equation 2.21. In this case P1 and P3 are the

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Table 6

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Rate equations for carbon-carbon dioxide reaction

Rate = $kP_{CO_2}^n$

Ref	k	n	Temp. Range °K	Particle Type	Remarks
71	1.35 exp (-16300/T _s) (g/cm ² .s.atm) 63.5 exp (-19500/T _s) (g/cm ² .s.atm)	0	1123 - 1223 1223 - 1573	graphite graphite	Experiment was conducted in pure CO ₂ using cylindrical particles. A first order reaction with respect to steam partial pressure was assumed. The rate equations given are based on the external surface area, and is the rate of surface reaction.
72	1.2 x 10 ⁻⁵ exp (-17600/T _s) (g/cm ² .s)	0	1013 - 1133		There were no indication to the type of carbon used in the experiment or the area to which the rate equation was based on. The rate equation given is the rate of the overall reaction.

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partial pressures of steam and P_2 is the partial pressure of hydrogen. Thus the order of the char-steam reaction varies with the steam partial pressure in a similar way to that in which the char-carbon dioxide reaction varies with carbon dioxide partial pressure as supported by Walker et al (41).

The retarding effect of hydrogen in the char-steam reaction has been reported, while there is still uncertainty about the effect of carbon monoxide with some data showing inhibition while the others show no effect at all on this reaction (54,59-61).

In a manner similar to carbon-carbon dioxide reaction, Reide (60) and Klei (61) concluded that under a typical gasification temperature, the overall rate of char-steam reaction is chemical reaction controlled and is appropriately described by the volumetric rate equation given by Equation 2.7. However these investigators were unable to correlate their experimental data based on this model but instead assumed the validity of surface reaction. Wen and Chaung (36) also assumed the surface reaction model to describe the char-steam reaction in their modelling of the entrained bed coal gasifier. Johnson (62) concluded that char-steam reaction should be described by the volumetric reaction and proposed the following equation to describe the rate equation:

$$\frac{dx}{dt} = f_L k_I (1 - x)^{2/3} \exp(-\alpha x^2)$$
(2.22)

where x = fraction of carbon conversion of char

$$f_{L} = f_{o} e^{(8467/T_{o})}$$

- f = relative reactivity factor of coal/char
- T_o = maximum temperature to which char has been exposed prior to gasification, °K

$$k_{I} = \frac{\exp(9.0201 - 31705/T) (1 - \frac{P_{CO} P_{H_{2}}}{P_{H_{2}O} K_{I}^{E}})}{[1 + \exp(-22.16 + 44787/T) (\frac{1}{P_{H_{2}O}} + 16.35 \frac{P_{H_{2}}}{P_{H_{2}O}} + 43.5 \frac{P_{CO}}{P_{H_{2}O}})]^{2}}$$

$$P_{CO}, P_{H_2}, P_{H_2O} = \text{partial pressures of CO}, H_2 \text{ and } H_2O$$

$$K_1^E = 10^{(7.49 - 7079/T)}$$

$$\alpha = \frac{52.5 P_{H_2}}{1 + 54.3 P_{H_2}} + \frac{0.521 P_{H_2}^{1/2} P_{H_2O}}{1 + 0.707 P_{H_2O} + 0.5 P_{H_2}^{1/2} P_{H_2O}}$$

Different values of the activation energy for the char-steam reaction have been reported in the literature which lie approximately in the range 45-65 kcal/mole. These values are close to those of the char-carbon dioxide reaction (55-65 kcal/mol). The rates of the char-carbon dioxide and char-steam reaction are typically in the same order of magnitude. This view has been supported by a number of investigators (22,30,41,54). Walker and Kini (22) suggested that the rate of the char-steam reaction is about three times that of the char-carbon dioxide reaction while Batchelder (54) reported that these two reactions have equal rates under a typical gasification temperature. A few of the char-steam rate equations are given in Table 7.

2.3.3. Gas-Gas Reactions

2.3.3.1. Water Gas Shift Reaction

The water gas reaction is the reaction between steam and carbon monoxide and can be represented as follows

$$CO + H_2O \neq CO_2 + H_2$$
 (2.23)

This homogeneous gas-gas reaction is slightly exothermic having heat of reaction of about -7.8 kcal/mol (63). According to Johnstone et al (64), the water gas reaction which is catalysed by the carbon surface and the impurities present, is fast and rapidly approaches equilibrium.

There are still however contradicting views reported on the mechanism of the water gas reaction. According to Lowry (11) this reaction does not take place in the gas phase but purely on the carbon surface, while Haslam (65) reported that to some extent the reaction does take place in the gas phase. Batchelder et al (54) found that the water gas reaction attained equilibrium much more rapidly at the carbon surface than in the gas phase indicating that there is a parallel gas phase reaction. Gwosdz (66) reported that the interaction of carbon monoxide and steam takes place on the carbon surface and the proportions of gaseous components produced correspond to the equilibrium established at the surface.

Different approaches in treating the water gas reaction have been

Table 7

Rate equations for carbon-steam reaction

Ref	k	Unit of Rate	n	Temp. Range °K	Particle Type	Remarks
60	k ₁ = 11.94 exp (-8238/T _s)	mo1/cm ² •s	1	773 - 1173	graphite	A graphite rod was used and placed in a gaseous mixture of $N_2 - H_20$ with steam partial pressure varied from 7-12 mm Hg. Rate equation given is the overall rate of reaction based on the external surface area. Effect of mass transfer was found to be small.
64	k ₁ =2 x 10 ⁸ exp (-32005/T _s)	mol/s•g of carbon	0.58	1033 - 1089	Activated carbon	The sample particle was placed in a gaseous mixture of argon and steam with mole fraction of steam varied from 0.107 - 0.534. The rate equation is the overall rate of reaction. However mass ransfer resistance was found to be small.

Rate = $kP_{H_20}^n$ or Rate = $k_1 C_{H_20}^n$

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Ref	k .	Unit of R	n	Temp. Range °K	Particle Type	Remarks .
73	$k = 3190 \exp (-25020/T_s)$ $k = 19.2 \exp (-17680/T_s)$	g/cm ² •s•atm g/cm ² •s•atm	1	1273 - 1433 1123 - 1273	graphite	Rate of reaction was determined in an environment of pure steam at 1 atm. First order reaction with steam partial pressure was assumed. The rate equations given are the rate of surface reaction based on the external surface area.
74	$k = 3.66 \times 10^{3} \exp (-26700/T_{s})$ k = 1.95 x 10 ⁹ exp (-41600/T_{s})	g/cm ² •s g/cm ² •s	0	1256 - 1589 1256 - 1589	petroleum coke electrode graphite	The rate equations are for the actual rate of reaction under the experimental condition based on the external surface area. However due to small particle sizes used $(63 - 74 \mu)$ the rate can be assumed to be the rate of surface reaction.

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used by many workers on gasifier modelling. The majority of them assumed that the reaction goes to equilibrium (36, 67), while the others (39) take into account the reaction kinetics. Purdy et al (68) found that the inclusion of the water gas reaction kinetics improved their prediction of the performance of the gasifier under study compared to the equilibrium assumption of the water gas reaction.

The equilibrium constant for the water gas reaction has been correlated by Parent and Katz (63) and follows the Van't Hoff Isochore relationship.

$$K_{\rm MG} = 0.0265 \exp (3955.7/T)$$
 (2.24)

The catalytic effect of impurities present in coal on the water gas reaction makes it difficult to establish a general rate equation for this reaction. Singh and Saraf (69) studied this reaction over an iron-base catalyst and proposed the following equation:

Rate =
$$F_w$$
 (2.77 x 10⁵) x (x_{CO} - x^{*}_{CO}) exp(-27760/1.987T) x
P^(0.5-P/250) exp(-8.91 + 5553/T) (g-mol/s of ash) (2.25)

where F_w = relative catalytic of ash to that of iron-base catalyst.

$$\mathbf{x}_{CO}^{*} = \frac{1}{P} \left[\frac{{}^{P}_{CO_{2}} {}^{P}_{H_{2}}}{{}^{K}_{ws} {}^{P}_{H_{2}}} \right]$$

P = total pressure, atm P_{CO_2} , P_{H_2} , P_{H_2O} = partial pressure of CO₂, H₂ and H₂O respectively, atm K = equilibrium constant for water gas reaction (eqn. 2.24)

2.3.3.2 Gaseous Combustion

A number of the gaseous components that are generated in a gasifier can undergo further combustion with oxygen to produce more heat. These include hydrogen, carbon monoxide and most of the components released due to pyrolysis.

The rates of gaseous combustion are much faster than those of the gas-solid reactions (36,42). According to Wen and Chaung (36) gaseous combustion can be considered instantaneous and therefore can be assumed to be completed as long as there is enough oxygen.

3. AIM AND SCOPE OF PRESENT WORK

All the previous studies on spouted bed coal gasification concentrated on assessing the performance of the system based on the overall conversion achieved from relatively small systems (13-15).

In order to improve the understanding of the spouted bed coal gasifier in general, it is necessary to obtain some information regarding the internal behaviour of the system which is expected to be related to the hydrodynamics of spouted beds. Previous studies on spouted beds (1) indicated that the bed hydrodynamics strongly depend on the column diameter. As it is very likely that large bed diameters would be employed if spouted bed gasifiers are to be chosen for commercial operations, the development of a reliable mathematical model is essential for design and scale-up purposes. It is therefore the aim of the present work to perform a study of the in-bed gas composition and temperature. In addition the theoretical model developed by Mathur and Lim (2,4) is applied to gasification and tested against the experimental data obtained.

Based on the observation reported by Rovero et al (7) regarding the existence of axial and radial profiles of gaseous components in their studies on the decomposition of ozone in a spouted bed and the RTD measurement of tracer gas made by Lim and Mathur (4), the following experimental measurements will be performed.

- axial and radial concentration profiles of gaseous components and gas temperature profile in the annulus.

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- axial concentration profiles of gaseous components and gas temperature profile in the spout.

The effect of the following operating variables on the resulting profiles will be studied:

- operating with a char bed as opposed to an inert bed

- gasification with air/steam mixtures

- gasification with oxygen/steam mixtures

- variation in types of coal fed to the gasifier

- bed height

- char rather than coal used as feed

Gaseous samples collected will be analysed for the following components: oxygen, hydrogen, methane, nitrogen, carbon dioxide and carbon monoxide.

4. EXPERIMENTAL APPARATUS

4.1 Gasification System

The existing 0.3 m diameter unit built under a contract awarded by Energy Research Laboratories, CANMET was used throughout the work. Detailed equipment description and designs are given elsewhere (75) and only a brief description will be presented here.

The schematic diagram of the system is given in Figure 4. Coal which is stored in a 45-gallon drum is fed to and metered through a vibra screw feeder and pneumatically transported using air or oxygen to either the top or the bottom of the bed. For this study only bottom feeding was used. In this case coal feeding into the bottom of the reactor is made through a 31.8 mm inlet orifice. The reactor is 0.5 m OD x 1.8 m high cylindrical column with a 60° conical base. To tolerate high gasification temperature the column is refractory-lined to an inside diameter of 0.305 m. Solid entrainment is reduced by provision of a 0.77 m ID x 0.62 m high disengaging section above the cylindrical section. The spouting medium which is also the gasification medium is injected into the reactor through the same coal inlet orifice.

Hot gaseous products with some entrained coal fines leave the top of the gasifier and pass through a series of two cyclones provided with char receivers at the bottom, three double pipe heat exchangers and finally to a knock-out drum and a dust filter before being burnt in the incinerator. In a number of selected runs, char collected from the first cyclone was recycled into the reactor via a rotary valve.

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Figure 4 - Schematic flow diagram for the gasification system.

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For start-up purposes a propane fired gas burner is provided in order to preheat the reactor to the desired experimental temperature.

4.2 Gas Sampling

A gas sampling probe was designed as shown in Figure 5. The probe consists of a 356 mm long x 6.35 mm OD SS 316 tube. Mounted at one end of the tube is a 12.7 mm OD porous stainless steel filter with an average pore opening of 90 μ . In order to be able to measure the gas temperature at the suction point a 1.59 mm (1/16") Chromel-Alumel thermocouple is inserted inside the tube. This arrangement is achieved by having a branch tee as shown in the diagram. A valve is incorporated so that the probe can be isolated from the sampling line whenever necessary.

The schematic diagram for the sampling line is shown in Figure 6. The gas sample drawn from the gasifier and which is free from any solid material due to the stainless steel filter mounted at the tip of the probe is passed through a knock-out drum where most of the condensed steam is removed. The dry gas leaving the knock-out drum is collected in a gas sampling tube fitted with two valves at both ends. A vacuum pump is used to suck the gas sample at a flowrate measured using a rotameter installed in the sampling line. A nitrogen line taken from a high pressure nitrogen cylinder is provided to clear off any possible blockage caused by the accumulation of coal dust on the stainless steel filter.

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Figure 6 - Schematic flow diagram for the gas sampling line.

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5. EXPERIMENTAL TECHNIQUE

5.1 Gasification Process

Detailed experimental techniques for the overall gasification process have been described by Watkinson et al (76) and only a brief description will be presented here.

A known amount of crushed coal or inert (depending on the bed material selected) was initially charged into the reactor and was then heated up using combustion products from the external nozzle mixed propane burner. In this case the gas from the burner formed the major portion of the spouting fluid. As soon as a temperature of about 450°C was attained in the bed, continuous coal feeding into the reactor was initiated, while the propane input was slowly reduced to zero. Due to the coal combustion inside the reactor, the bed temperature continued to rise to the desired value and the steam flow was turned on. The air to coal ratio was then adjusted and the system was allowed to reach a steady state which normally took about one hour after feed adjustments before any gas sampling could be performed. The gasification experiments were carried out by Mr. Gordon Cheng and Mr. Sam Low as part of the requirements of the research contract.

5.2 Gas Sampling

Six different axial positions and three different radial positions corresponding to each level have been chosen for this study. This required six sampling probes, one to be placed at each level.

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These various positions are shown schematically in Figure 7.

An isokinetic gas sampling technique was used throughout this work. Hence at a particular location, the gases velocities inside the sampling probe and that inside the gasifier should be equal. Each of these velocities and therefore the actual volumetric gas flowrate to be drawn were theoretically determined. The Mamuro-Hattori equation (1) was used to determine the axial gas flow distribution in the annulus, while the axial flow distribution in the spout was obtained from a mass balance as described by Mathur and Lim (2). For this purpose the McNab equation (1) was used to calculate the average spout diameter.

The gas sampled from a chosen position was first allowed to pass through the sampling line for about 8 min before it was collected and isolated from the line by closing the two valves at the ends of the gas sampling tube. Typically it took about 2-3 min to purge out the whole sampling line completely. Meanwhile, the actual gas temperature was recorded at every 2 min interval. The sample collected was subsequently stored in a pressure-lock gas syringe for analysis at a later time. While working on a particular probe, all the other five remaining were pulled out to positions such that their tips would remain inside the gasifier wall. This procedure was practised to minimise the disturbance of the gas flow path which would have been caused by the presence of these probes. There was no set sequence for any particular sampling position to be chosen. This was done at random.

In order to construct the average gas temperature profiles in the gasifier, instantaneous gas temperatures at all the 18 locations

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No.	z (cm)	r (cm)	No.	z (cm)	r (cm)	No.	z (cm)	r (cm)
6A	65	11.4	4A	45	11.4	2A	25	11.4
6B	65	6.3	4B	45	6.3	2B	25	6.3
6C	65	0	4C	45	0	2C	25	0
5A	55	11.4	3A	35	11.4	1A	8.5	5.0
5B	55	6.3	3B	35	6.3	1B	9.1	4.0
5C	55	0	3C	35	0	1C	11.4	0

Figure 7 - Exact locations of sampling points in the bed.

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involved were taken almost instantaneously. Three different sets of readings, taken at the beginning, in the middle and at the end of the sampling process were recorded and averaged.

5.3 Blockage of Sampling Probes

Blockage of the gas sampling probes was experienced; however this was not frequent. The main reason for the blockage was the accumulation of coal fines on the filter elements mounted at the tips, and these were removed by flushing with nitrogen. In order to efficiently flush away these particles, the whole sampling line was isolated beforehand by closing the isolating valve as indicated in Figure 6.

5.4 Gas Analysis

The gas analysis was performed using a Hewlett-Packard 5710A gas chromatograph. The unit was equipped with a thermal conductivity detector and an automatic integration system. Two different columns were used to separate the gaseous components. These were a 3.2 mm x 2.13 m molecular sieve A column to separate hydrogen, oxygen, nitrogen, methane and carbon monoxide and a 3.2 mm x 3.96 m Porapak Q column to separate carbon dioxide. Throughout the analysis both the oven and the detector were maintained at temperatures 80°C and 200°C respectively.

To calibrate the G.C. five gas samples having known quantities of different species were injected into the system. A calibration table as shown typically in Appendix C will then be generated, which will be used as the internal standard. Before performing any analysis a standard gas sample was first analysed to ensure that the calibration is correct.

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6. MODEL DEVELOPMENT AND SIMULATION

Steady state models of char gasification in a spouted bed blown with air/oxygen and steam have been developed based on the general one-dimensional and the streamtube spouted bed models proposed by Lim and Mathur (2,4) and the gasification model of Yoshida and Kunii (76) which they applied to the fluidized bed. In both cases isothermal gasifiers have been assumed.

6.1 Development of Model

6.1.1 Dominant Reactions

The following reactions have been considered to describe the gasification process

gasification:

$$C + H_2O + CO + H_2$$
 (6.1)

$$C + CO_2 \rightarrow 2CO \tag{6.2}$$

combustion:

$$C + O_2 \rightarrow CO_2 \tag{6.3}$$

$$CO + 1/2 O_2 \rightarrow CO_2$$
 (6.4)

$$H_2 + 1/2 \ 0_2 \rightarrow H_2 0$$
 (6.5)

water gas shift:

$$CO + H_2 O \rightleftharpoons CO_2 + H_2 \tag{6.6}$$

Following the correlation proposed by Wen and Lee (19) to determine the product distribution of char combustion, carbon dioxide has been assumed to be the only product (Equation 6.3). The water gas shift reaction was

assumed to be in equilibrium throughout the bed.

At a typical gasification temperature and atmospheric pressure, the rate of reaction between hydrogen and char was found to be insignificant (28-29). This reaction has been ignored in the present model.

6.1.2 Reaction Kinetics

A simple reaction model based on the particle geometric surface area has been used to describe reactions 6.1 - 6.3. In this case the resistances due to gas transport in the boundary layer and in the ash layer have been ignored and the rate is assumed to depend on temperature and the partial pressure of reacting species given as follows:

Rate =
$$k_{overall} \exp(-E/RT)P_g^n$$
 (6.7)

The different kinetic parameters chosen for this simulation are given in Table 8.

The rates of gas phase combustion given by equation 6.4 - 6.5 are normally fast compared to char combustion as concluded by Wen and Chaung (36). These rates of reaction are usually high and can be considered to be instantaneous. Hence these reactions were assumed complete as long as there was oxygen existing.

6.1.3 Bed Hydrodynamics

The presently available correlations on the hydrodynamics of spouted beds have been used for this purpose, with the assumption of

Table 8

Kinetic parameters used in gasification modelling

A. Reaction kinetic

*Rate	-	k overall _i	$exp(-E_i/RT)$	x ⁿ i
-------	---	---------------------------	----------------	------------------

Reaction	^k overall _i	E _i /R	×i .	n	Unit of Rate	Ref.
Char-02	8710	18000	P ₀₂ (atm)	1	g/s cm ²	48
Char-CO ₂	4×10^8	29790	C _{CO2} (mol/m ³)	1	mol/m^2	30
**Char-H ₂ 0	1.2×10^9	2 979 0	$C_{\rm H_{2}O}$ (mol/m ³)	1	$mo1/m^2$	

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X.

B. Equilibrium constant for water-gas shift reaction (63)

 $K_{ws} = 0.0265 \exp(3955/T)$

*Based on external suface area

**Assuming rate constant of C/H_2O reaction is three times the rate constant for the C/CO_2 reaction.

their validity at high temperatures. These different correlations are given in Table 9.

6.1.4 One-Dimensional Model

This model assumed plug flow of gas both in the spout and in the annulus.

In developing the equations describing the gasification process in spouted bed, the procedure proposed by Yoshida and Kunii (76), has been adopted. Using this method the change in gas concentration due to gas-solid and gas-gas reaction was accounted for separately.



6.1.4.1 Reactions in the Spout

A. Changes due to gas-solid reactions

A balance over spout height ΔZ for component i gives

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Tab	le	9

Hydrodynamic parameters for model

Parameter	Equation	Ref.
Maximum spoutable bed height	$H_{\rm m} = 0.105 d_{\rm c} (d_{\rm c}/d_{\rm p})^{0.75} (d_{\rm c}/d_{\rm i})^{0.4} \lambda^2 / \rho_{\rm p}^{1.2}$	1
Annulus gas velocity (superficial)	$U_a = 0.88 U_{mf} (1 - (1 - z/H_m)^3)$	5
Minimum spouting velocity	$U_{ms} = (d_p/d_c) (d_1/d_c)^{1/3} \sqrt{\frac{2g H(\rho_p - \rho_f)}{\rho_f}}$	1
Mean spout diameter	$D_{s} = 0.118 G^{0.49} d_{c}^{0.68} / \rho_{b}^{0.41}$	1
Annulus voidage	$\varepsilon_a = 0.43$	
Spout voidage	$\varepsilon_s = 1 - 0.2 z/H$	6
Spout gas velocity (superficial)	$\overline{U}_{s} = [U_{s} A_{c} - U_{a}(A_{c} - A_{s})]/A_{s}$	
Minimum fludization velocity	$U_{mf} = 0.5 (U_{bf} + U_{tf})$	77
Velocity beginning of fluidization	$(\text{Re})_{\text{bf}} = [(18.1)^2 + 0.0192 \text{ Ga}]^{0.5} - 18.1$	77
Velocity total fluidization	$(\text{Re})_{\text{tf}} = [(24)^2 + 0.0546 \text{ Ga}]^{-0.5} - 24$	77

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$$\overline{U}_{s} A_{s} C_{si} = [A_{s} \overline{U}_{s} + d(\overline{U}_{s} A_{s})] [C_{si} + d C_{si}] + U_{r} \pi D_{s} C_{si} dz + \sum_{i} R_{j} A_{s} (1 - \varepsilon_{s}) dz$$
(6.8)

where R_j , j = 1,2,3 are the rate of reactions represented by equations 6.1 - 6.3 respectively expressed as kmol/(m³ of particle volume s). Ignoring any change in volumetric flux due to changes in molar flux and pressure:

$$U_{r} = -\frac{1}{\pi D_{s}} \frac{d}{dz} (\overline{U}_{s} A_{s})$$
(6.9)

Combining Equation 6.8 and 6.9 and assuming a constant spout diameter, gives the following simplified equations for individual components:

$$\frac{dC_{s1}}{dz} = \frac{-R_1(1 - \varepsilon_s)}{\overline{U}_s}$$

$$\frac{dC_{s2}}{dz} = \frac{(R_1 + 2R_2)(1 - \varepsilon_s)}{\overline{U}_s}$$
(6.10)
$$\frac{dC_{s3}}{dz} = \frac{R_1(1 - \varepsilon_s)}{\overline{U}_s}$$

$$\frac{\mathrm{dC}_{\mathrm{s4}}}{\mathrm{dz}} = \frac{(\mathrm{R}_3 - \mathrm{R}_2) (1 - \varepsilon_{\mathrm{s}})}{\overline{\mathrm{U}}_{\mathrm{s}}}$$

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$$\frac{dC_{s5}}{dz} = \frac{-R_3(1 - \varepsilon_s)}{\overline{U}_s}$$

$$\frac{dC}{dz} = 0$$

Equation 6.10 can be rewritten to give the following expression for the change in component concentration ΔC_{si} over bed height Δz resulting from gas-solid reaction

$$\Delta C_{s1} = \frac{-R_1 (1 - \varepsilon_s)}{\overline{U}_s} \Delta z \qquad (6.11)$$

$$\Delta C_{s2} = \frac{(R_1 + 2R_2)(1 - \varepsilon_s)}{\overline{U}_s} \Delta z \qquad (6.12)$$

$$\Delta C_{s3} = \frac{R_1 (1 - \varepsilon_s)}{\overline{U}_s} \Delta z \qquad (6.13)$$

$$\Delta C_{s4} = \frac{(R_3 - R_2)(1 - \varepsilon_s)}{\overline{U}_s} \Delta z \qquad (6.14)$$

$$\Delta C_{s5} = \frac{-R_3(1 - \varepsilon_s)}{\overline{U}_s} \Delta z \qquad (6.15)$$

$$\Delta C_{s6} = 0 \tag{6.16}$$

Thus the component concentration C'_{si} resulting from gas-solid reactions are given by:

$$C_{s1}' = C_{s1} + \Delta C_{s1}$$

$$C_{s2}' = C_{s2} + \Delta C_{s2}$$

$$C_{s3}' = C_{s3} + \Delta C_{s3}$$

$$C_{s4}' = C_{s4} + \Delta C_{s4}$$

$$C_{s5}' = C_{s5} + \Delta C_{s5}$$

$$C_{s6}' = C_{s6}$$
(6.17)

B. Changes Due to gaseous combustion

On the assumptions that the gaseous combustion proceeds instantaneously, and as long as there is oxygen present the change in component concentrations $\Delta C'_{si}$ due to gaseous combustions is given by

$$\Delta C_{s1}' = C_{s3}'$$

$$\Delta C_{s2}' = -C_{s2}'$$

$$\Delta C_{s3}' = -C_{s3}'$$
(6.18)
$$\Delta C_{s4}' = C_{s2}'$$

$$\Delta C_{s5}' = -(C_{s2}' + C_{s3}')$$
$$\Delta C_{s6}' = 0$$

Combining Equations 6.17 and 6.18 gives the component concentration C_{si} " after taking into consideration both the gas-solid reaction and the gaseous combustion which can be represented as follows:

$$C_{s1}^{"} = C_{s1} + \Delta C_{s1} + C_{s3} + \Delta C_{s3}$$

$$C_{s2}^{"} = 0$$

$$C_{s3}^{"} = 0$$

$$C_{s4}^{"} = C_{s4} + \Delta C_{s4} + C_{s2} + \Delta C_{s2}$$

$$C_{s5}^{"} = C_{s5} + \Delta C_{s5} - C_{s3} - \Delta C_{s3} - C_{s2} - \Delta C_{s2}$$

$$C_{s6}^{"} = C_{s6}$$
(6.19)

According to Yoshida and Kunii (76) who modelled fluidized bed coal gasification, the conversion predicted by taking into consideration gaseous combustion was very close to conversion where such reactions are neglected. The importance of considering such reactions in spouted bed char gasification modelling will be discussed later.

C. Equilibrium of Water Gas Shift Reaction

If ΔC_s is the individual change in molar concentration of H_2O , CO, H_2 and CO₂ due to water gas shift reaction, Yoshida and Kunii (76) state that the following relationship must be satisfied.

$$K_{ws} = \frac{(C_{s4}" + \Delta C_{s}) (C_{s3}" + \Delta C_{s})}{(C_{s2}" - \Delta C_{s}) (C_{s1}" - \Delta C_{s})}$$

or
$$\Delta C_{s} = \frac{(K_{ws} C_{s2} C_{s1}) - (C_{s4} C_{s3})}{C_{s4} + C_{s3} + K_{ws} C_{s2} + K_{ws} C_{s1}}$$
 (6.20)

D. Equations Describing Reaction in the Spout

In the situation where C_{s2} " and C_{s3} " equal zero, Equation 6.20 is not applicable and the final component concentration C_{si} ''' at height $z + \Delta z$ equals C_{si} " represented by Equation 6.19. On the other hand if the gaseous combustion is neglected or when there is no oxygen ... present, the final concentration C_{si} ''' at height $z + \Delta z$ can be obtained by combining Equations 6.17 and 6.20:

$$C_{s1}''' = C_{s1} + \Delta C_{s1} - \Delta C_{s}$$

$$C_{s2}''' = C_{s2} + \Delta C_{s2} - \Delta C_{s}$$

$$C_{s3}''' = C_{s3} + \Delta C_{s3} + \Delta C_{s}$$

$$(6.21)$$

$$C_{s4}''' = C_{s4} + \Delta C_{s4} + \Delta C_{s}$$

$$C_{s5}''' = C_{s5} + \Delta C_{s5}$$

 $C_{s6}''' = C_{s6}$

6.1.4.2 Reactions in the Annulus

A. Changes Due to Gas-Solid Reactions

A balance over the annulus height Δz for component i gives

$$U_{a}A_{a}C_{ai} + U_{r}\pi D_{s}C_{si}dz = [A_{a}U_{a} + d(A_{a}U_{a})] [C_{ai} + dC_{ai}] + \Sigma R_{j}A_{a} (1 - \varepsilon_{a}) dz \qquad (6.22)$$

where U_r is given by

$$U_{r} = \frac{1}{\pi D_{s}} \frac{d}{dz} (U_{a}A_{a})$$
(6.23)

Combining 6.22 and 6.23 and rewriting the resulting equations as the change in concentration ΔC_{ai} over the annulus height $z + \Delta z$ gives

$$\Delta C_{a1} = \left[\frac{-R_1(1-\epsilon_a)}{U_a} + \frac{(C_{s1} - C_{a1})}{U_a}\frac{dU_a}{dz} + \frac{(C_{s1} - C_{a1})}{A_a}\frac{dA_a}{dz}\right] \Delta z \qquad (6.24)$$

$$\Delta C_{a2} = \left[\frac{(R_1 + 2R_2)(1 - \epsilon_a)}{U_a} + \frac{(C_{s2} - C_{a2})}{U_a}\frac{dU_a}{dz} + \frac{(C_{s2} - C_{a2})}{A_a}\frac{dA_a}{dz}\right] \Delta z \quad (6.25)$$

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$$\Delta C_{a3} = \left[\frac{R_1(1-\epsilon_a)}{U_a} + \frac{(C_{s3} - C_{a3})}{U_a}\frac{dU_a}{dz} + \frac{(C_{s3} - C_{a3})}{A_a}\frac{dA_a}{dz}\right] \Delta z \qquad (6.26)$$

$$\Delta C_{a4} = \left[\frac{(R_3 - R_2)(1 - \epsilon_a)}{U_a} + \frac{(C_{s4} - C_{a4})}{U_a}\frac{dU_a}{dz} + \frac{(C_{s4} - C_{a4})}{A_{a}}\frac{dA_a}{dz}\right] \Delta z \quad (6.27)$$

$$\Delta C_{a5} = \left[\frac{-R_3(1-\epsilon_a)}{U_a} + \frac{(C_{s5} - C_{a5})}{U_a}\frac{dU_a}{dz} + \frac{(C_{s5} - C_{a5})}{A_a}\frac{dA_a}{dz}\right] \Delta z \qquad (6.28)$$

$$\Delta C_{a6} = 0.$$
 (6.29)

B. <u>Changes Due to Gaseous combustion and Water Gas Shift Equilibrium</u> and Equations Describing Reactions in the Annulus

Following the same procedure previously adopted, the expressions for ΔC_a due to gaseous combustion, water gas shift reaction and the final molar concentration at annulus height $z + \Delta z$ were obtained which were in the same form as Equations 6.19 - 6.21, but at conditions referring to those in the annulus.

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6.1.5 Streamtube Model



In this model as proposed by Lim and Mathur (4) vertical plug flow of gas in the spout and plug flow along curved streamlines in the annulus have been assumed. No mixing or dispersion was allowed in the direction of flow and normal to the streamlines.

6.1.5.1 Gas Flow Path

To determine the gas flow path in the annulus, grid points were constructed by dividing the bed into M equal vertical intervals and the annulus width at the top into N equal horizontal intervals with each interval representing one flow path. Letting Q(J-1) be the volumetric flow between streamlines J-1 and J, taking overall gas mass balance at any level I gives

$$Q(J-1) = U_{a}(I)\pi[R(J-1, I)^{2} - R(J,I)^{2}]$$
(6.30)

where $U_a(I)$ is the superficial annulus gas velocity at level I and R(J,I) specifies the ordinate of the streamline J at level I (the column axis is taken as the reference). The first streamline (J=1) was assumed to coincide with the column wall. Letting the interval between streamlines J and J+1 be the flow path J, the superficial gas velocity $U_{az}(I)$ along flow path J was calculated using the relationship.

$$U_{az}(I) = U_{a}(I)/\cos \theta_{a}$$
(6.31)

In this model θ_a was taken to be the angle formed between the line joining the centre of flow path J at levels I and I-1 and the horizontal. Denoting PL(I) as the distance between these two centre points, the total path length for flow path J from the spout-annulus interface to the top of the bed at I=M was given by Σ PL(I). 6.1.5.2 Reactions in the Spout

Since the same assumptions were made with regard to the gas flow in the spout as in the one-dimensional model, the reactions in the spout were described by the same equations, i.e. equations $6.17^{-} - 6.21$.

6.1.5.3 Reactions in the Annulus

A. Changes Due to Gas-Solid Reactions



A component mass balance over length ΔL along the flow path J gives

$$U_{az}A_{az}C_{azi} = [A_{az}U_{az} + d(A_{az}U_{az})][C_{azi} + dC_{azi}] + \sum_{j} R_{j} A_{az}(1 - \varepsilon_{az}) dL$$
(6.32)

On the assumption of no change in volumetric flux due to the change in molar flux and pressure, Equation 6.32 reduces to the following expressions for the change in the individual component concentration ΔC_{azi} over the path lengh ΔL due to the gas-solid reactions:

$$\Delta C_{azl} = \frac{-R_1(1 - \varepsilon_{az})}{U_{az}} \Delta L$$
 (6.33)

$$\Delta C_{az2} = \frac{(R_1 + 2R_2)(1 - \varepsilon_{az})}{U_{az}} \Delta L \qquad (6.34)$$

$$\Delta C_{az3} = \frac{R_1 (1 - \varepsilon_{az})}{U_{az}} \Delta L$$
 (6.35)

$$\Delta C_{az4} = \frac{(R_3 - R_2)(1 - \varepsilon_{az})}{U_{az}} \Delta L \qquad (6.36)$$

$$\Delta C_{az5} = \frac{-R_3(1 - \epsilon_{az})}{U_{az}} \Delta L$$
 (6.37)

$$\Delta C_{az6} = 0 \tag{6.38}$$

In this case $\varepsilon_{az} = \varepsilon_{a}$.

B. Changes Due to Gaseous Combustion and Water Gas Shift Equilibrium

Following the same procedure adopted in all the previous cases, the expressions for ΔC_{az} due to the water gas shift equilibrium and the final component concentrations at the path length L + Δ L were derived and were in the same form given by Equations 6.18 - 6.20 but at conditions referring to those in the flow path J.

6.1.6 Outlet Component Gas Concentration

The final component gas concentration leaving the gasifier was calculated by combining the streams leaving each region. Thus the exit gas concentration C_{ei} for component i can be expressed as

One-dimensional model:

$$C_{ei} = \frac{U_{aH} A_a C_{aHi} + \overline{U}_{sH} A_s C_{sHi}}{U_s A_c}$$
(6.39)

Streamtube model:

$$C_{ei} = \begin{bmatrix} \Sigma & U_{aH} & A_{azHj} & C_{azHij} \end{bmatrix} + \overline{U}_{sH} & A_{s} & C_{sHi} \\ \underline{j=1} & U_{s}A_{c} \end{bmatrix}$$
(6.40)

Any reaction in the fountain has been ignored.

6.2 Calculation Procedure

The equations describing the change in gas concentration due to the gas solid reactions given by Equations 6.10 - 6.15 for the spout, Equations 6.24 - 6.28 for the annulus in the one dimensional model and Equations 6.33 - 6.37 for the flow path J in the streamtube model are non-linear differential equations and were solved using the 4th order Runge-Kutta technique. The component concentrations in the one dimensional model were obtained by alternately solving the equations for the spout and the annular region over any incremental height Δz . Both of these regions have identical initial boundary conditions which were equal to the inlet gas component concentration.

For the streamtube model, the gas concentration profiles in the spout were initially obtained using the inlet gas concentration as the boundary conditions. The gas concentrations along the gas flow path J were then solved using the gas concentration in the spout taken at the mid point between streamlines J and the J + l as the inlet condition.

6.3 Model Simulation and Discussion

A number of simulated runs were performed using both the one-dimensional and the streamtube models to predict the axial and the radial gas composition profiles as well as the overall conversion achieved from a spouted bed coal gasifier. The column geometry used in the experimental system has been chosen for this purpose, using air/oxygen and steam as the only gasification medium. In addition, the importance of some operating variables such as the total bed height and the bed temperature in determining the gasifier performance was also investigated.

6.3.1 Axial Gas Composition Profiles

Figure 8 shows the axial gas composition profiles in the annulus predicted using the one-dimensional model. For both carbon monoxide and hydrogen, the gas composition increases with height, reaching a maximum

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Figure 8 - Axial gas composition profiles in the annulus predicted using the one-dimensional model.

value in the conical region and reduces to a minimum at the conecylinder intersection. The concentration of combustible species increases steadily thereafter with the carbon monoxide showing a sharper increase than the hydrogen. The C-CO2 reaction was partly responsible for this sharp increase in carbon monoxide as a function of height. The low H_2/CO ratio predicted throughout the bed was due to the small amount of steam used in the feed, i.e. air/steam (w/w) = 8. These proportions of H₂ and CO would actually become closer to each other if the steam/ oxygen ratio in the feed stream was higher. This is clearly shown in Figure 9 which gives the axial gas composition profiles in a spouted bed gasifier blown with oxygen/steam. In this case an oxygen/steam (w/w) ratio of unity was used as the feed. The appearance of maximum and minimum values for these gases was due to the change in the annulus area along the bed height in the conical region. The combined effect of the variation in annulus area A_a and the annulus superficial velocity U_a with bed height causes equations 6.25 and 6.26 which described the change in carbon monoxide and hydrogen concentration to change sign as the calculation proceeds from the bed inlet to the cone-cylinder intersection. On the other hand the carbon dioxide composition shows a maximum value at the cone-cylinder interface and steadily decreasing values thereafter. The sharp increase in carbon dioxide close to the bed inlet was due to the product of combustion as shown by a sudden diminishing of oxygen. Within this bed height the term R3 in Equation 6.27 becomes dominant to give a positive increase in carbon dioxide concentration. The oxygen concentration falls to zero in that first few centimeters. Once the oxygen concentration diminishes R3 becomes zero

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Figure 9 - Axial gas composition profiles in the annulus predicted using the one-dimensional model blown with oxygen rather than air.

and further increase in heights cause the carbon dioxide concentration to decrease.

The corresponding spout gas composition profiles are shown in Figure 10. In contrast to those in the annulus there is hardly any reaction taking place in this region except for some combustion. The oxygen content thus remains high. The main reason for the absence of any significant reactions is the high gas velocity in the spout, which results in a very small change in the components' concentration along the spout height as represented by Equation 6.10. The spout voidage has no significant effect in determining the gas concentration profiles in the annulus. Using an average constant value of 0.65, a value measured by Lim (3) at z = H, produced an almost indistinguishable result from the one shown in Figure 10. Rovero et al (7) made the same conclusion regarding the effect of voidage on conversion in the spout.

In order to investigate the importance of carbon monoxide and hydrogen oxidation in the modelling of spouted bed char gasification, a run was simulated in which these two reactions were ignored. In comparing the results, no significant differences could be detected. There was hardly any reaction between steam and carbon dioxide with char in the spout and the char oxidation in the annulus was almost instantaneous in utilizing the oxygen as soon as it enters the reactor. Hence carbon monoxide and hydrogen never coexist with oxygen anywhere in the gasifier and very little error would be introduced by ignoring the combustion of carbon monoxide and hydrogen in models where radial mixing is assumed not to be present.



Figure 10 - Axial gas composition profiles in the spout predicted using the one-dimensional model.

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The total bed height was found to have no significant effect on the axial gas composition profiles. As shown in Table 9 all of the correlations used in predicting the bed hydrodynamics except for the spout voidage and the minimum spouting velocity were independent of the total bed height. As has already been shown the effect of spout voidage on conversion was negligible, while a constant overal gas mass flowrate rather than a constant U_s/U_{ms} ratio has been used in the prediction. However, under a given operating condition U_{ms} was used as a check to ensure that the ratio U_s/U_{ms} is greater than 1.

Figure 11 shows the axial profiles for carbon monoxide along the different flow path predicted using the streamtube model. For all the flow paths, the gas composition rises steadily along the path length from the spout-annulus interface to the top of the bed, with the one closest to the wall increasing most sharply and then levelling off as it reaches the bed top. For comparison the average profile predicted by the one-dimensional model was also plotted. The only difference that can be observed is the absence of any maximum/minimum value in the conical region for the streamtube model as compared to the predictions of the one-dimensional model. This difference is attributed to the independence of the equations describing the change in the gas concentration along the path length in the annulus area A_a (Eqn. 6.33 - 6.37).

Plotted in Figure 12 are the axial carbon monoxide and hydrogen composition profiles along the centre of the annulus, obtained using the one-dimensional and the streamtube models. The predictions from both models are relatively close except for the lower section of the bed.

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Figure 11 - Carbon monoxide profile along streamlines in the annulus predicted using the streamtube model.



6.3.2 Radial Gas Composition Profiles

Figure 13 shows the radial gas composition profiles at the top of the bed predicted using the streamtube model. As expected more carbon monoxide and hydrogen are found close to the column wall due to the longer path length and gas residence time in the corresponding flow paths. Meanwhile the carbon dioxide composition rises steadily from its original value in the spout to a maximum and decreases steadily towards the column wall. This maximum value, located close to the spout-annulus interface is due to the char combustion as the gas from the spout entered the corresponding flow path. At the same time the oxygen composition is reduced to zero.

Figure 14 shows the radial carbon monoxide composition profiles at different levels in the bed which can be deduced directly by following a vertical straight line drawn at any bed level z in Figure 11. As shown there is a large variation in gas composition along the middle section of annulus.

In contrast to the streamtube model, the one dimensional model fails to give any radial variation in the gas composition profiles as shown in Figure 15 for carbon monoxide and hydrogen. This is not surprising as uniform radial gas concentration has been one of the assumptions made in the initial model development.

6.3.3 Exit Gas Composition

In predicting the exit gas composition the spout gas and the annulus gas have simply been added, assuming that no reaction occurs in the freeboard. Obviously in a reactor operating at 1073 K, reaction



Figure 13 - Radial gas composition profiles at the top of the bed predicted using the streamtube model.



Figure 14 - Radial carbon monoxide composition the annulus at various bed levels predicted using the streamtube model.



Figure 15 - Comparison of the radial hydrogen and carbon monoxide profiles in the annulus at the top of the bed; ----, streamtube model;----, one-dimensional model.

would occur. These gas-gas reactions in the fountain will be considered later in Chapter 8, where comparison will be made between the predicted and the experimental results.

6.3.3.1 Effect of Total Bed Height

Figure 16 gives the variation in the component gas composition as a function of the total bed height predicted using the one-dimensional model. The proportions of the carbon monoxide and hydrogen in the exit stream increase as the total bed height increases, while that of oxygen steadily decreases. As discussed earlier and from Figures 8 and 10, all of the carbon monoxide and hydrogen in the product stream is due to the reaction in the annulus. All of the oxygen in the annulus is consumed almost instantaneously, while in the spout there is insufficient reaction to consume the oxygen. High total bed height results in high carbon dioxide concentration in the spout at the top of the bed and low concentration in the annulus measured at the same level as shown in Figures 8 and 10. The combination of these two streams results in the observed carbon dioxide concentration profiles in which carbon dioxide increases to a maximum and then decreases thereafter as the total bed height is increased. Hence improved gas quality from a spouted bed gasifier would be obtained as the total bed height increased. The gasifier would give the best performance when operated at its maximum spoutable height. Theoretical investigation by Littman et al (6) on catalytic reaction in the spouted bed gave the same conclusion with regard to the effect of the total bed height on the overall conversion.



Figure 16 - Effect of the total bed height on the exit gas composition predicted using the one-dimensional model. Assume no reaction of spout and annulus gas. Overall volumetric gas flow is kept constant.





Under the same operating conditions the variation in the exit gas composition with the total bed height was also predicted based on the streamtube model. No significant differences between these two models could be detected as shown in Figure 17, which gives the comparison for carbon monoxide and hydrogen.

6.3.3.2 Effect of the Air/Steam Ratio

Figure 18 shows the variation in the product gas distribution as a function of the inlet air/steam ratio predicted using the one-dimensional model. In general much higher percentage of carbon monoxide and hydrogen and hence improved gas quantity could be obtained at low air/steam ratios. Increasing the air/steam ratio causes both the hydrogen and carbon monoxide to decrease with the hydrogen composition falling off more sharply than that of the carbon monoxide. Operating at low air/steam would give a high proportion of hydrogen in the product gas. On the other hand fuel gas richer in carbon monoxide would be produced by maintaining high air/steam ratios. Little variation in carbon dioxide composition was observed within the range investigated. The oxygen fraction increases slightly as the air/steam ratio increases, since more unreacted oxygen in the spout will leave the gasifier. As in the previous case, predictions using the streamtube model gave a trend undistinguishable from the simpler model.

In practice it is very difficult to operate at a low air/steam ratio in order to maintain a reasonably high gasification temperature, as very little oxygen is available for combustion. Extra heat can, of course, be supplied by recirculating hot solids from an external

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Figure 18 - Effect of air/steam ratio on the exit gas composition predicted using the one-dimensional model. Assume no reaction between spout and annulus gas.

combustor, but here we are considering only systems blown with gases containing oxygen. For these systems when oxygen is used instead of air, a high proportion of steam is required to control the temperature, particularly if operation in the dry-ash mode is desired.

6.3.3.3 Effect of the Average Bed Temperature

Figure 19 shows the variation of the component gas composition as a function of the average bed temperature again assuming simple mixing of spout and annulus gas without reaction. As expected on increasing the average bed temperature, the proportion of carbon monoxide and hydrogen in the product stream increases. This was due to the strong dependence of both the $C-CO_2$ and the $C-H_2O$ reactions on the reaction temperature. The decrease in the oxygen composition at high reaction temperature was due to the increase in the extent of combustion in the spout to give a high carbon dioxide composition in this region. At the same time higher temperature results in a much lower carbon dioxide composition in the annulus. The combination of these two streams at the top of the bed lead to the observed carbon dioxide profile, showing a minimum value at around 820°C and a slight increase as temperatures However at a much higher temperature when all of the oxygen increase. has been consumed, a decrease in the carbon dioxide composition is expected. As in all earlier cases, both the streamtube and the one-dimensional models gave the same prediction.

6.3.3.4 Effect of Particle Size

Figure 20 shows the variation in the overall gas composition as a function of particle diameter.



Figure 19 - Effect of the average bed temperature on the exit gas composition predicted using the one-dimensional model. Assume no reaction between spout and annulus gas.



Figure 20 - Effect of the particle size on the exit gas composition predicted using the one-dimensional model. Assume no no reaction between spout and annulus gas. Overall volumetric gas flow is kept constant.

As shown in Table 9 particle diameter is an important variable which will affect most of the hydrodynamic properties of a spouted bed. In general, under the same gas spouting flowrate a large particle diameter will give a high gas velocity in the annulus and hence a much lower velocity in the spout than a smaller particle. Thus an improved reaction in the annulus is normally obtained from small size particles while large particles result in an increase in the extent of conversion in the spout. Combining these two streams gives the effect of the particle size on the overall gas composition as shown in Figure 20. The carbon monoxide increases steadily to show a maximum value at the particle diameter 2.5 mm and decreases slightly thereafter. No such pattern is observed for all the other gas components. Prediction using the streamtube model shows similar variation in the overall gas composition as a function of particle size.

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7. EXPERIMENTAL RESULTS AND DISCUSSION

Gas composition profile measurements were attempted in over eighteen runs of the gasifier. Western Canadian coals were used: sub-bituminous Forestburg coal and bituminous Balmer coal. Both the proximate and ultimate analysis on these two coals are given in Table 10. About ten of the attempted runs were successful in that complete data were collected for the given modes of operation and operating conditions. The coal particle size used throughout the present study is -3.36 + 1.19 mm.

The present work is part of an ongoing research programme on coal gasification in spouted beds which has been in progress over the past five years. All the profile measurements were made during runs performed by G. Cheng and S. Low who are looking into various aspects of gasification. Thus to avoid any confusion on the data compiled the run numbers of Cheng and Low have been adopted in this work.

In presenting the results all gas compositions are expressed on a moisture-free basis. The axial gas concentration profiles are presented for three different radial positions in the bed referred to here as positions A, B and C. The exact locations of these three different positions in the bed are given in Figure 7 — position A is nearest the wall, and position C is on the centre-line of the reactor.

7.1 General Considerations

7.1.1 Sampling Rate

Throughout the present work the intention was to sample the gas

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Properties of coals tested

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Ultimate Analysis (% wt MAF)	Forestburg	Balmer
C	74.87	88.71
Н	4.44	4.86
S	0.64	0.21
N	1.59	1.14
0	18.46	5.07
- Proximate Analysis		· · · · · · ·
% Moisture	23.71	1.03
% Volatiles	29.44	20.93
% Fixed Carbon	33.25	66.67
% Ash	13.60	11.37
Free Swelling Index	1	1.5

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isokinetically. Due to the variations in gas composition and temperature from point to point in the bed and from one run to another, an assumed gaseous environment and bed temperature have been chosen in order to estimate the gas sampling rate (Appendix A). Therefore some variation between the estimated sampling velocity and the actual gas velocity in the gasifer at the point of suction is expected to exist. However from the gas analyses given in Table 11 which were collected from point 2A (Figure 7) in Run 89 at three different flowrates, the effect of sampling rate on the gas composition seems to be very small even at four times the estimated isokinetic rate. In the worst case the present estimated rate can only be a few percent in excess of the actual isokinetic value. Thus it can be concluded that the procedure adopted in estimating the isokinetic sampling rates from different positions in the bed can be used without introducing any significant error in the results obtained.

7.1.2 Drying

No steam was added into the system when gasifying Forestburg coal since this coal as fed contained 24% moisture. Thus all of the steam available for the reaction must have been released from the fresh coal particles as they dried. According to the measurements made by Lim (3) using a particle size and column diameter relatively close to the one employed in the present study, the particle velocity in the spout ranges from 3 m/s at the bottom to 0.8 m/s at the top of a 0.9m bed. Based on these values and for a bed of 1 m high, the average residence time of

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| Gas
Component | u /*u
s iso | | | |
|------------------|----------------|------------------------|--------|--|
| | 1 | 3 | 4 | |
| | · · · · | Gas Composition, Vol % | (dry) | |
| Н | 30.307 | 31.131 | 31.379 | |
| CO ₂ | 8.872 | 8.103 | 8.242 | |
| 02 | 0.109 | 0.09 | 0.167 | |
| N ₂ | 28.44 | 29.086 | 28.854 | |
| CH4 | 3.428 | 3.347 | 3.110 | |
| CO | 28.844 | 28.235 | 28.249 | |

Table 11

Effect of sampling rate on gas composition

*Estimated as in Appendix A.

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a freshly fed coal particle in the spout will be in the order of one second. To estimate the amount of water released within this time due to drying, the equation proposed by McIntosh (78) can be used:

$$t = \frac{L\rho_{wc}[MC_{o}/(1 + MC_{o})] d_{p}}{h(T_{g} - T_{wb}) 2} \times \left[\frac{1}{3}(1 - Bi)(1 - \frac{MC}{MC_{o}}) + \frac{Bi}{2}(1 - (\frac{MC}{MC_{o}})^{2/3})\right]$$
(7.1)

where Bi = Biot number, $\frac{hd_p}{2k_c}$

t = time, s
h = heat transfer coefficient,
$$W/m^2 \cdot {}^{\circ}K$$

d_p = particle diameter, m
k_c = thermal conductivity of coal, $W/{}^{\circ}K.m$
L = latent heat of vaporization of water, J/kg
MC = coal moisture content at time t (fraction, dry basis)
MC₀ = MC at t = 0
T_{wb} = adiabatic wet bulb temperature, ${}^{\circ}K$
T_g = gas temperature, ${}^{\circ}K$
 P_{wc} = density of raw coal, kg/m³

The heat transfer coefficient in the spout can be estimated from the following equation (1)

$$Nu = \frac{2}{[1 - (1 - \varepsilon)^{1/3}]} + \frac{2}{3\varepsilon} Pr^{1/3} Re^{0.55}$$
(7.2)

where ε in the spout can be taken as 0.85(3), and the Nusselt and Reynolds numbers are based on particle diameter. For the present case using coal of average particle size of 2.1 mm and an average temperature of 800°C in the spout, the average heat transfer coefficient in this region has been estimated to be 736 W/m².°K. Using an estimated value of 0.069 W/m.°K for the thermal conductivity of coal (79), calculations suggest that about 30% of the water remains inside the coal particles as it leaves the spout. Thus the assumption of complete drying within the spout may not be justified for relatively large coal particles or when a shallow bed is used.

7.1.3 Pyrolysis

The rate of pyrolysis which is known to be much slower than drying depends on a number of factors including the particle temperature. On the assumption of no radial temperature gradient within the particle, the temperature T_p at any time t attained by a particle placed in a gaseous environment of temperature T_b can be deduced from the following unsteady state equation:

$$\frac{T_{p} - T_{p0}}{T_{b} - T_{p0}} = 1 - \exp\left[\frac{-hA_{p}t}{m_{p}Cp}\right]$$
(7.3)

where T_{po} is the initial particle temperature. Under the same operating conditions discussed in section 7.1.2 the same coal particle would leave the spout with an average particle temperature of 450°C taking T_{po} as 25°C. Applying equations 2.1 and 2.2 to describe the rate of pyrolysis and taking the values of K_0 ' and E' as 0.019s⁻¹ and 19.72 kJ/mol respectively for Forestburg coal (79) it would appear that only about 0.1% of the volatiles in the fresh feed coal are released in the spout. As the particles will only spend a very brief period in the fountain (\approx 0.1 s) most of the pyrolysis will take place in the annulus where the particles residence time will be much longer.

According to Al-Jarallah (79) there is a substantial radial temperature gradient within a 2.3 mm coal particle during the heating up period. In this case devolatilisation in the spout will be much less than what has been estimated.

7.2 Forestburg Coal-Air

7.2.1 Axial Gas Composition Profiles

A. Gas Composition Along Position B

Figure 21 shows the axial gas composition profiles taken along position B which is roughly 9 cm from the reactor wall. As shown the oxygen concentration drops almost instantaneously to essentially zero as soon as it enters the gasifier due to the high rate of char combustion. The decrease in the oxygen concentration is accompanied by a sharp increase to a maximum value in the carbon dioxide concentration indicating that carbon dioxide forms the major product of combustion. Whether there is any carbon monoxide produced in addition to carbon dioxide as a result of char combustion could not be confirmed in this study. The drop in carbon dioxide composition in the conical region which is in line with the model prediction as shown in Figure 8 can be explained by the C-CO₂ reactions. In addition, the effect of gas

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Figure 21 - Axial gas composition profiles along position B for Forestburg coal-air system (Run 93).

circulation in this region (8) may have also contributed to the drop in concentration. However, contrary to the theoretical prediction which shows a continuous decrease in the carbon dioxide composition throughout the bed, the experimental profile rises again from a value of 11.4% at a height of 50 cm. Beyond this level the composition remains relatively constant. The existence of such a profile towards the top of the bed is thought to be due to the competition between the amount released by pyrolysis and that consumed by the C-CO₂ reaction. As discussed earlier, the fresh feed coal that enters the annulus from the top is still in the initial stages of pyrolysis. Thus there are both pyrolysis and gasification reactions in the upper section of the annulus. Recent work on pyrolysis (79) showed that a high yield of carbon dioxide (15% w/w MAF coal) could be obtained by pyrolysing Forestburg coal at 600°C.

Both the carbon monoxide and hydrogen show similar profiles with the gas composition increasing along the bed height. The existence of points of inflection in the composition curves at the bed height of 25 cm which is close to the cone/cylinder intersection (at z = 20 cm) may have been the effect of gas recirculation in the conical region. Relatively similar profiles for these two components have also been predicted for this region solely from a theoretical approach as is shown in Figure 8. In this case maximum and minimum peaks were shown which were purely due to the column geometry for this lower section of the bed as has already been discussed in the previous chapter. Presumably the observed profiles occur not only because of gas recirculation but also are to some extent due to the conical bottom employed. The gasification

reactions must have been responsible for the increase in both carbon monoxide and hydrogen composition towards the top of the annulus. However, based on a simulated model using the operating conditions employed in this run as the input, gasification alone would have not resulted in the relatively equal proportion of hydrogen and carbon monoxide measured throughout the bed. Possibly in addition to gasification there are other sources of hydrogen and carbon monoxide which may be significant. Pyrolysis could be one of these sources. The H_2/CO ratio released during pyrolysis depends very much on temperature with more carbon monoxide being released at low temperature. As much as 5% (w/w MAF Coal) of carbon monoxide could be obtained at 600°C while the corresponding figure for hydrogen is 1% (79). Due to the large amount of volatiles present in the Forestburg coal, reforming reactions of these volatiles with steam probably occurs. Taking carbon, hydrogen and oxygen as the only components present in the volatiles, the following basic reaction can be assumed:

$$C_{m}H_{n} 0 + (m - 1) H_{2}0 + mC0 + (\frac{n}{2} + m - 1) H_{2}$$
 (7.4)

The values of m and n for Forestburg coal can be estimated from the data reported by Al-Jarallah (79). A simple calculation performed to estimate the amount of carbon monoxide and hydrogen released separately by reforming reactions pyrolysis and gasification showed that all of these reactions are about equally important as sources of carbon monoxide and hydrogen in the bed. Particle segregation may also play an

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Gas yield due to pyrolysis at 600°C for Forestburg coal and Balmer coal (79)

Coal	Gas Yield, % (w/w) MAF coal			
	H ₂	CO	CO ₂	СНц
Forestburg	1.2	5.0	15.0	3.7
Balmer	1.0	0	0	5.3

important role especially in the upper section of bed, where improved gas-solid reactions are achieved from smaller size particles. The coal as fed contains particles from 1.19 mm to 3.36 mm. Furthermore the particles in the bed will keep reducing in size as the reactions proceed until they are entrained away. In such a system where particles of different sizes exist, the smaller sizes tend to congregate near the top of the bed (80).

Low methane composition throughout the bed has been observed increasing from zero at the inlet to only about 3.5% at z = 65 cm. This low value is expected as all of the methane in the bed comes from pyrolysis rather than the C-H₂ reaction which is insignificant under the present condition.

B. Gas Composition Along Position A

Figure 22 shows the gas composition profiles along position A which is close to the column wall. There is no significant difference with regard to both the oxygen and methane composition profiles to those previously observed along position B.

Both the hydrogen and carbon monoxide show similar concentration profiles, increasing smoothly from zero at the inlet to about 23% at z = 65 cm. As discussed earlier the high concentrations at the upper section of the bed for these two components may be due to pyrolysis and reforming reaction in addition to the normal gas-solid reactions. However, in contrast to the profiles along position B, no point of inflection along the concentration curves is observed in the conical



Figure 22 - Axial gas composition profiles along position A for the Forestburg coal-air system (Run 93).

region. A similar profile for carbon monoxide concentration has been predicted from the theoretical approach using the streamtube model as shown in Figure 12. Thus it is possible that the actual gas flow pattern along this position is similar to that assumed in the stream tube model, i.e. plug flow of gas from inlet orifice to the top of bed. For the case of position B which is close to the annulus centre the plug flow pattern in the conical region could not exist due to gas recirculation. Presumably this effect is less experienced along this lower section of position A due to its close location to the wall, thus allowing the plug flow pattern of gas to be maintained.

In general the carbon dioxide composition profile shows a pattern similar to that observed along position B. As argued in the previous case, the high carbon dioxide concentration at the top of the bed may be due to the additional amount released from pyrolysis. The sudden jump in the concentration between the bed level z = 25 cm and z = 35 cm is partly due to the effect of char recycling which will be discussed in the next section.

C. Gas Composition along position C

Figure 23 shows the gas composition profiles taken along the spout.

Both the carbon monoxide and hydrogen show similar profiles with very low concentrations at the lower section of the bed and then increase steadily to about 13% at the bed height z = 65 cm. The high concentrations observed in the upper section of the column contradict

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Figure 23 - Axial gas composition profiles along position C for the Forestburg coal-air system (Run 93).

the model predictions as shown in Figure 10. According to the model, both of these components should not be present anywhere in the spout. The high compositions measured may have been contributed by the volatiles. However from what has been discussed above the fresh feed coal in the spout is only in the very initial stage of pyrolysis and it is very unlikely that they will release any substantial amount of volatiles. Another source of volatiles in the upper section of the spout is the particles which emerge from the annulus which are still undergoing pyrolysis. These particles which cross-flow into the spout in the upper section of the bed normally spend very little time in the inner upper part of the annulus. Due to segregation these particles are usually large in size, in which case pyrolysis can last for several seconds.

As in the annulus, the methane composition remains relatively low throughout the bed. A value of about 1.8% has been measured at the bed level z = 65 cm. It is believed that the presence of methane in this region as well as in the annulus is solely due to pyrolysis. Judging from its profile in Figure 23 and Figures 21 and 22 for the annulus region, it is evident that there is still coal pyrolysis taking place even in the upper section of the bed.

The oxygen concentration curves shows both a minimum and maximum peak within the conical region before finally reducing to zero. The total disappearance of oxygen may have not been entirely due to char combustion as shown by the model prediction in Figure 10. Possibly it is the combination of char, volatiles and gaseous combustion that

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consumes all of the available oxygen in the region. Interestingly the carbon dioxide profile traces a curve which turns out to be the mirror image of the oxygen profile. Its high concentration in the upper section of the bed is mainly due to the product of combustion and may be to some extent contributions from pyrolysis and reforming reactions. The fluctuations in both the carbon dioxide and oxygen composition profiles in the conical region are due to combustion and gas recirculation.

Throughout this work it has been assumed that the spout in the system is stable in the vertical position and its centre coincides with the centre of the column. All the samples taken along the column axis are assumed to represent those in the spout. Piccinini et al (5) however observed that the spout in their system tended to wander, resulting in poor reproducibility of results for the region. Unfortunately, no such observation can be made here. Comparing the measured composition profiles obtained along this region for all the runs, relatively similar shapes have been maintained by all the gas components. From this observation it can be concluded that those assumptions made earlier are valid.

The gas composition profiles in the conical region as clearly shown in Figures 21-23 have been repeatedly observed in all the runs. Thus experimental errors should be ruled out with regard to the measured experimental points at the different bed levels in the conical region.

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7.2.2 Radial Gas Composition Profiles

Plotted in Figure 24 are the radial gas composition profiles in the annulus for carbon dioxide and carbon monoxide taken at different bed levels for Run 93. As shown there is a variation in the radial component gas composition within the annulus itself, which becomes more significant as one goes deeper into the bed. In terms of the models discussed, the existence of such a radial variation can only be explained by assuming the validity of steamtube model to describe the gas flow pattern in the annulus.

7.2.3 Effect of Operating Condition

7.2.3.1 Char Recycle

In order to study the effect of the recycle of char from the external cyclone on the gas composition profiles experimental runs using both modes of operation, i.e. with and without char recycling, were performed. Char re-enters the bed at a single point in the annulus and is conveyed with a small flow of air. Plotted in Figure 25 are the measured profiles along position A obtained from Run 90 (char recycling) and Run 89 (no char recycling). In this case a direct comparison can be made to distinguish any differences that might exist. It is clearly shown from these plots that the gas composition curve traces by each component except carbon dioxide show similar shapes. As far as these components are concerned, no differences can be detected to discriminate these two modes of operation. The differences in the actual composition values are due to the slight variation in the operating variables



Figure 24 - Radial gas composition profiles for Forestburg coal-air system (Run 93): a) carbon dioxide, b) carbon monoxide.



Figure 25 - Effect of char recycling on composition profiles for the Forestburg coal-air system: a) Run 90 (recycling), b) Run 89 (no recycling).

employed, as it was not possible to maintain identical conditions in both cases. The only distinguishable trend which can differentiate the two systems is shown by the carbon dioxide composition profile. The sudden increase in the carbon dioxide concentration between the bed level z = 25 cm and z = 35 cm is partly due to the contribution from the char combustion with the incoming air which is used to transport the recycling char. For this system the recycling inlet to the reactor is located close to the bed position z = 35 cm. This sudden increase in carbon dioxide between the two bed levels has also been observed in all runs operated with char recycling. Only a gradual increase in the carbon dioxide composition between these two bed levels is observed in runs where no char recycling are employed as is shown in Figure 25b for Run 89. There are, however, no differences shown on the profiles along position B and C. This could be due to the location in the bed from which the samples were taken. In this case they are further away from the column wall and the char recycling inlet.

The decline in the component gas compositions beyond the bed height H in Run 89 is considered to be due to gas mixing in the fountain. The same factor is also responsible for the decline observed in Run 90. However, judging from the position at which the drop in gas compositions took place, higher bed height should have actually been employed in Run 90. The indicated bed height in Figure 25 as well as in all other figures was actually taken at the end of the operation. In a number of occasions, a slight reduction in the bed height during the course of the experiment was experienced.

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7.2.3.2 Bed Height

Plotted in Figure 26 are the axial gas composition profiles along position B taken for Runs 91 and 93, which were operated under different total bed heights as indicated.

The same general trends in the gas composition profiles are still being maintained in both cases except for carbon dioxide where a minor difference exists. In this case the drop in the composition in the upper section of the conical region as shown in Run 93 is no longer observed when operated at a lower bed height as in Run 91. Instead the carbon dioxide composition increases continuously throughout the bed. As discussed earlier, the effect of pyrolysis can be significant depending on the location in the bed. Since some of the particles entering the annulus are still undergoing pyrolysis, the top of the bed can be affected most by this process. Thus the increase in the carbon dioxide composition throughout the conical region in run 91, which is in this case located close to the bed top, is probably due to the contribution from pyrolysis, rather than experimental error since the same trends are also observed in most other runs operated under relatively deep and shallow beds respectively.

As shown in Figure 26b the increase in the gas composition in the fountain is mainly due to gas mixing beyond the bed as evident by the corresponding drop in the profiles along position A. In addition there is also some contribution from pyrolysis. The extent of gas-solid reaction in the fountain is not known and its contribution to the change in gas concentration in the region cannot be assessed.

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The gas composition profiles along position A are similar to those observed in Run 90 as illustrated in Figure 24a. A comparison of Figure 24a and Figure 21 shows no major difference in the concentration curves within the bed between these two cases. The same conclusion can also be made with regard to the profiles along position C.

As in the previous case, this comparative study is based on the results obtained from two systems operated under similar rather than identical operating conditions. Thus, as expected there exists some variation in the actual gas composition at any given position.

7.2.3.3 Average Bed Temperature

The effect of average bed temperature can be illustrated by comparing Figure 25a and Figure 22 to give the composition profiles along position A for Run 91 and 93 which were operated at average bed temperature of 800°C and 909°C respectively. As concluded earlier these profiles as well as those along positions B and C show no major differences. Within this temperature range the C-H₂O and C-CO₂ reactions are still low and the change in the carbon monoxide, hydrogen and carbon dioxide concentrations due to these reactions are still insignificant to be noticed. Relatively similar gas heating values have been produced by gasifying Forestburg coal between temperatures 790 and 930°C. (13)

7.3 Forestburg Coal - Oxygen

In this study the effect of using oxygen rather than air on the gas profiles in the gasifier was investigated. There was however a small

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flow of air added to the system through the feed line to transport the fresh feed coal into the gasifier. To control the gasification temperature a large amount of steam has to be added to the system to give H_2O/O_2 ratio in the order of 3.45 (w/w). Only one set of readings on the gas composition profiles was successfully obtained under this operating condition. Hence the effect of other operating variables could not be examined.

7.3.1 Axial Gas Composition Profiles

A. Gas Composition along position B

Plotted in Figure 27 are the gas composition profiles along position B. Comparison with Figure 21 where air was used instead shows that there are major differences in the gas composition pattern between these two systems. Both the carbon dioxide and hydrogen profiles show very little change throughout the bed. The concentrations of CO, CO_2 and H₂ increase sharply within a few centimeters from the inlet orifice in the lower section of the conical region and remain relatively constant thereafter. The slight increase followed by a continuous decrease in the hydrogen composition in the fountain is due to gas mixing in the region as evident by the shape of the corresponding hydrogen composition profiles along positions A and C (Figures 28 and 29 respectively). The same factor is also responsible for the observed carbon monoxide profiles in the fountain. A large ratio of H₂/CO is consistently produced throughout the bed, which is due to a high



Figure 27 - Axial gas composition profiles for Forestburg coal-oxygen system along position B (Run 96).

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 $H_20/0_2$ ratio in the feed.

As in the previous system the sharp reduction in the oxygen concentration is accompanied by a rapid increase to a maximum value in the carbon dioxide concentration followed by a continuous decrease throughout the bed. The C/CO_2 reaction is partly responsible for this decrease. Due to mixing the gas concentration rises again in the fountain as shown. No methane was found in the bed. It may have been burned away as soon as it was released.

A slightly higher proportion of hydrogen as compared to carbon monoxide has been predicted using the char gasification model. This is not in a very good agreement with the experimental data. An estimate of the amount of carbon monoxide and hydrogen released individually from the volatiles reforming, pyrolysis and gasification indicated that the effects of pyrolysis may not be very significant when compared to gasification and reforming reactions. From this argument, it is possible that reforming reaction may be an important potential source of carbon monoxide and hydrogen in addition to gasification reactions.

B. Gas Composition along positions A and C

Figure 28 shows the gas composition profiles along postion A. In general these profiles are similar to those along position B. The trends shown in the bed can be explained in the same manner as discussed above while those in the fountain are possibly due to gas mixing.

Figure 29 shows the gas composition profiles along position C at the reactor centre line. As in the case of the Forestburg coal-air

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Figure 28 - Axial gas composition profiles for Forestburg coal-oxygen system along position A (Run 96).



Figure 29 - Axial gas composition profiles for Forestburg coal-oxygen system along position C (Run 96).

system the profiles along this position are not in accordance with theoretical predictions for the spout where only oxygen and carbon dioxide are supposed to be present. Comparing this plot and those in Figures 27 and 28 shows that their shapes are relatively similar. Based on this comparison it is likely that these profiles do not represent those in the spout, in which case the spout might have wandered off the column axis under this operating condition. However, no confirmation can be made at this stage as there are no other data available under this operating condition.

7.4 Balmer Coal - Oxygen

Four different runs were attempted to gasify Balmer Coal using oxygen and steam. However most of the runs lasted for about two hours except Run 103 where a total running time of three hours was achieved. Longer running times were prevented by a pressure build-up in the gasifier, in which case the system had to be shut down immediately. Hence only one set of readings taken from Run 103 will be discussed in this section. Even for this run, complete results could not be presented as there was no time to collect any samples along position C (spout). Due to some modifications carried out on the gasifier samples from the conical region could not be collected.

7.4.1 Axial Gas Composition Profiles

Plotted in Figure 30 are the gas composition profiles observed along positions A and B respectively. Comparing these two plots with

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Figure 30 - Axial gas composition profiles for Balmer coal-oxygen system in Run103: a) Along position A, b) Along position B.

those in Figures 27 and 28, i.e. the composition profiles obtained from the Forestburg coal-oxygen system clearly shows that the composition curves in the bed traced by all the components are relatively similar. The difference in the actual composition values are due to the differences in the coals used and to different operating conditions used, mainly the average bed temperature. For this run the average bed temperature was 913°C compared to 830°C in Run 96.

As in all the previous cases, the sharp decrease in the oxygen composition is expected which is due to combustion, while all the methane present in the bed which shows a maximum composition of about 2% comes from pyrolysis.

The steady rise in the carbon monoxide and hydrogen compositions throughout the bed along both positions A and B are due to gasification reactions and to some extent the volatiles reforming. As is evident by the methane composition profile, there is still pyrolysis taking place in the lower section of the bed. An estimate on the amount of hydrogen released due to pyrolysis showed that its contribution to the observed hydrogen profile was not significant, and certainly was insignificant in the case of carbon monoxide. This coal as fed contains about 21% volatiles which is about half of that in Forestburg coal. Recent attempt to pyrolyse this coal showed that no carbon monoxide and carbon dioxide were released mainly due to its low oxygen content. In addition about 5.3% (MAF Coal) of methane were obtained at 600°C as compared to 1% for hydrogen (79). From this observation, it can be argued that the role of pyrolysis in determining the gas composition profiles in the bed

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may not be very important for this system.

The continuous decrease in the carbon dioxide composition along both positions was partly due to $C-CO_2$ reaction. As mentioned above there was no contribution from pyrolysis.

As in the case of the Forestburg coal-oxygen system the high H_2/CO ratio observed throughout the bed is due to the high H_2O/O_2 ratio in the feed. In this case a ratio of 2.87 (w/w) has been used.

7.5 Partially Devolatilised Forestburg Char-Air

A run was performed (Run 106) using partially devolatilised Forestburg char as the feed in order to examine the differences between coal and char as feed material. For this purpose the same operating conditions as in Run 93 were chosen so that direct comparison of the measured composition profiles between these two cases can be made. However there were still some variations in the actual conditions employed. Air/fixed carbon and steam/fixed carbon ratios of 4.6 and 0.723 respectively have been used in this run. The corresponding figures for Run 93 are 5.4 and 0.713. Thus an average bed temperature of 900°C has been achieved in this run compared to 800°C for Run 93.

The char sample was prepared in different batches by heating Forestburg coal over a period of 2 - 3 hours at a temperature of 400 - 500 °C. A spouted bed combustor equipped with an external gas burner was used for this purpose. The combustion products of propane and air from an external burner was used as the heating medium. To avoid any combustion of coal, the propane/air ratio entering the burner was carefully controlled so that no oxygen was present in the product stream leaving the burner. The properties of this sample are given in Table 13.

As in the case of the Balmer-oxygen system, gas samples from the conical region could not be collected.

7.5.1 Axial Gas Composition Profiles

Plotted in Figures 31 and 32 are the gas composition profiles along positions A, B and C. The penetration of oxygen into the bed is much deeper in this case as shown by the axial oxygen composition profiles. This observation strongly suggests that the rate of char combustion in this system is lower. Thus the drop in both hydrogen and carbon monoxide compositions along the bed is due to gaseous combustion. According to Johnson (62) exposing char samples to a high temperature prior to gasification reduces its reactivity. However this finding could not be confirmed in this study.

A higher proportion of carbon monoxide as compared to H_2 was measured throughout the bed. This is in contrast to the profiles observed in Run 93 as shown in Figures 21 - 23. This is an important difference between these two systems as the effects of operating conditions should be ruled out. However more experimental data are required to confirm these profiles.

7.6 Average Temperature Profiles

During the course of the experiments three different sets of

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Table 13

Forestburg	Partially Devolatilised Forestburg Char
74.87	84.71
4.44	2.55
0.64	0.51
1.59	1.80
18.46	10.43
	·
23.71	1.64
29.44	15.64
33.25	65.60
	Forestburg 74.87 4.44 0.64 1.59 18.46 23.71 29.44 33.25

13.60

% Ash

17.12

Properties of Forestburg coal and the partially devolatilised Forestburg char



Figure 31 - Axial gas composition profiles along position A for partially devolatilised Forestburg char-air system (Run 106).



Figure 32 - Axial gas composition profiles for partially devolatilised Forestburg char-air (Run 106): a) Along position B, b) Along position C.

instantaneous temperature readings at all of the points involved were recorded. These readings, which were taken at the beginning, in the middle and at the end of the experiment were then averaged to give the average axial temperature profiles along positions A, B, and C.

7.6.1 Forestburg Coal-Air

Figure 33 shows the average temperature profiles along positions A, B, and C taken for Run 93.

There exists a large variation in the gas temperature along the spout referred to here as position C. The rapid rise from 700° C at the bed level z = 25 cm to about 800° C at the bed level z = 35 cm is due to combustion as evident by the rapid drop in the oxygen concentration within the same bed levels shown in Figure 23. It is the combination of combustion and low incoming gas and solid temperatures that results in a very large temperature difference in the spout.

The temperature profile along position B which is close to the annulus centre is similar to that observed in a moving bed gasifier. In this case the combustion reaction is responsible for the high temperature at the bottom while the drop in temperature towards the top is due to gasification reactions.

The temperature profile along position A increases steadily to maximum value in the lower half of the column and reduces thereafter. This low temperature at the bottom is the result of the endothermic $C-H_2O$ and $C-CO_2$ reactions. The slight increase in values observed may have been due to the inflow of heat from the neighbouring region which is partly shown by the simultaneous decrease in temperatures along position B.



Figure 33 - Axial temperature profiles for Forestburg coal-air system (Run 93) along positions: \bigcirc , A; \Box , B; \triangle , C .
In addition to the axial variation in gas temperatures there are also significant radial temperature gradients, not only between the spout and annulus, but also within the annulus itself, especially in the lower section of the bed.

7.6.2 Forestburg Coal-Oxygen

The average temperature profiles along positions A, B and C taken for Run 94 are plotted in Figure 34.

As in the previous case the rapid increase in temperture along the spout is due to the heat released from combustion. According to Figure 29, there is still a substantial amount of oxygen along the centre of the column in the fountain. Hence combustion must also be responsible for the sharp increase in the gas temperature observed along this region.

Both the temperature profiles along positions A and B show similar trends increasing throughout the bed as well as in the fountain. The steady increase in temperature in the fountain is the result of gas mixing within the region to give a higher average gas temperature than that in the annulus. A very shallow bed was employed in this run. Thus it is possible that the continuous increase in temperature along the bed in the annulus is due to the higher temperature of the incoming solids from the fountain rather than combustion. Judging from the oxygen concentration along positions A and B as shown typically in Figures 27 and 28, it is unlikely that any combustion takes place in the upper section of the bed.



Figure 34 - Axial temperature profiles for Forestburg coal-oxygen (Run 94) along positions: (), A; (), B; \triangle , C .

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7.6.3 Partially Devolatilised Forestburg Char-Air

Figure 35 shows the average axial temperature profiles along positions A, B and C. Judging from the amount of oxygen in the bed as indicated in Figures 31 and 32, it is evident that the increase in temperature along the bed and in the fountain is due to combustion.



Figure 35 - Axial temperature profiles for partially devolatilised Forestburg char-air (Run 106) along positions: \bigcirc , A; \Box , B; \triangle , C .

8. COMPARISON BETWEEN MODEL AND EXPERIMENT

A model simulation of Run 93 has been performed and compared with the experimental data, based on the following assumptions in addition to those mentioned earlier in Chapter 6.

- 1. Effect of char recycling is neglected.
- 2. The gasification reactions and pyrolysis are treated separately. The gaseous products due to these two processes are combined in the fountain. The gas phase in this region is assumed to be perfectly mixed.
- 3. Gas phase combustion is assumed to continue in the fountains, as long as there is oxygen present. The gas components leaving the gasifier are in equilibrium with respect to the water gas shift reaction.

8.1 Estimation of Pyrolysis Product Distribution

The composition of the product of pyrolysis (weight fraction) can be estimated from the following equations proposed by Rejan (81)

$$CH_4 = 0.201 - 0.469 X_{vm} + 0.241 X_{vm}$$
 (8.1)

$$H_2 = 0.157 - 0.868 X_{vm} + 1.388 X_{vm}^2$$
 (8.2)

$$CO_2 = 0.135 - 0.900 X_{vm} + 1.906 X_{vm}^2$$
 (8.3)

$$CO = 0.428 - 2.653 X_{vm} + 4.845 X_{vm}^2$$
(8.4)

$$H_2 0 = 0.409 - 2.389 X_{vm} + 4.554 X_{vm}^2$$
 (8.5)

$$Tar = -0.325 + 7.279 X_{vm} - 12.880 X_{vm}^2$$
(8.6)

8.2 Gaseous Combustion in the Fountain

Only the combustion of carbon monoxide and hydrogen given by Equations 6.4 and 6.5 respectively are considered to be significant. These reactions are assumed complete as long as there is oxygen existing. Methane oxidation can be neglected as its concentration in the fountain, which solely comes from pyrolysis is generally low. The proportions of oxygen shared between these two reactions are estimated from the ratio of their rates of reaction. According to Sundaresan and Amundson (58) the combustion rates of carbon monoxide (R_A) and hydrogen (R_B) can be represented as follows:

$$R_{A} = k_{A} C_{CO} C_{O_{2}}^{1/2}$$
(8.7)

$$R_{B} = k_{B} C_{H_{2}} C_{O_{2}}^{1/2}$$
(8.8)

Thus the ratio ${\tt R}_A\!:\!{\tt R}_B$ or ${\tt R}_{AB}$ is given by

$$R_{AB} = \frac{k_A C_{CO}}{k_B C_{H_2}}$$
(8.9)

From their analysis which is applicable for reactions occurring in an environment of high oxygen concentration, the value of k_B is very much bigger than k_A . Under the present operating conditions, the oxygen concentration in the fountain is always low and it is very unlikely that the proposed rate constants are still valid. Due to the lack of other data, an assumed value of 1 has been assigned to the ratio $k_A:k_B$. Thus Equation (8.9) reduces to

$$R_{AB} = \frac{C_{CO}}{C_{H_2}}$$
(8.10)

8.3 Kinetic Parameters

The kinetic parameters for both types of coal under study are not available. Thus the same parameters as given in Table 8 have been chosen for this purpose. In this case the comparison will only focus on the trends predicted by the theoretical model and that obtained from experimental measurements rather than the actual values.

8.4 Axial Gas Composition Profiles

8.4.1 Profiles in the Annulus

Plotted in Figure 36 are the profiles for carbon dioxide, carbon monoxide and hydrogen predicted using the streamtube model and those measured along positions A and B. The predicted trends for both carbon monoxide and hydrogen are in line with the measured profiles where the gas composition increase continuously along the bed. The effect of pyrolysis could not be included in the model since there are no - 139 -



Figure 36 - Comparison between experimental and predicted axial profiles for Run 93 (Forestburg coal-air) using the streamtube model: a) Along position A, b) Along position B.

mathematical expressions to describe the particle movements in the bed. In addition the possibility of volatiles reforming in the system was not considered. As discussed earlier the inclusion of these two factors may be essential in order to accurately predict the hydrogen and carbon monoxide composition profiles in the bed. The predicted carbon dioxide profiles are not in agreement with that experimentally measured. Pyrolysis may be partly responsible for this difference.

Unlike the streamtube model, the one-dimensional model can only predict the average gas composition profiles, which is assumed to be the profile along the centre of the annulus. Exact comparisons could not be made here as no experimental data along the annulus centre were taken. However as already been discussed in Chapter 6, the profiles along the centre of the annulus predicted by both models (Figure 12) are relatively close giving a higher proportion of carbon monoxide than hydrogen throughout the bed.

The predicted and measured oxygen concentrations are in good agreement, and both drop sharply to zero very close to the bed inlet. This is due to the very high combustion rate compared to the other reactions. The C-H₂ reaction has been ignored in this model as this reaction has been assumed negligible under the present operating conditions.

In addition to the contribution from pyrolysis and to some extent the volatiles reforming reactions, there are several other factors which may have resulted in the difference between the predicted and measured profiles. The rate equations which describe reactions 6.1 - 6.3 were assumed to follow a surface reaction kinetic model. Under the typical gasification temperature and the relatively coarse particle size employed in the present study the gasification reactions have been found to take place simultaneously on the inside as well as on the outside surface of the particle (30). Thus these reactions may be more appropriately represented by the volumetric reaction model, in which the rate strongly depend on the fractional carbon conversion in the particle since the inside pore structure changes as the reactions proceed. Unfortunately this reaction model could not be applied here because the particle movements could not be described. There are also some errors introduced in predicting the profiles within the conical region by ignoring the effect of gas recirculation in the region.

In all the simulations performed, an isothermal gasifier has been assumed. Judging from Figure 33 such an assumption is justified especially when dealing with the annulus since the measured temperature is relatively constant to within about 50°C.

8.4.2 Profiles in the Spout

The difference between the predicted and the measured profiles in the spout can be obtained by comparing Figures 10 and 23. There is still a substantial amount of oxygen predicted in the spout which has not been observed experimentally. In addition large proportions of carbon monoxide and hydrogen have been measured in the upper section of the spout which are not in accordance with the predicted trends. Thus it appears that the contributions from pyrolysis and volatiles reforming play a major role in determining the axial gas composition profiles in the spout.

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8.5 Radial Gas Composition Profiles

Figure 37 shows the comparison between the predicted and measured radial composition profiles of carbon dioxide and carbon monoxide at z = 65 cm. As shown experimentally, there is a radial variation in the gas composition within the annulus. Thus in this respect the streamtube model will be superior to the one-dimensional model since it is able to predict radial composition profiles over the entire annulus, eventhough the predicted gradients were much too steep when compared to the actual measured values. It is likely that an improved prediction could be obtained by allowing some form of gas dispersion normal to the streamlines, which was not included in the present model.

8.6 Exit Gas Composition

The predicted exit gas compositions are not in good agreement with the experimentally determined data as shown by the plot in Figure 38. In this case the one dimensional model has been used. The same distributions would have also been obtained from the streamtube model. The similarity between these two models in this aspect has been discussed previously in Chapter 6. The observed variations between the predicted and the experimental values are directly related to the model's inability to closely predict the gas composition profiles within the bed.

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Figure 37 - Comparison between experimental and predicted radial carbon monoxide and carbon dioxide profiles for Run 93 (Forestburg coal-air):---, one-dimensional modal;---, streamtube model.



Figure 38 - Comparison between the experimental and predicted exit gas composition using the one-dimensional model for Run 93 (Forestburg coal-air).

9 CONCLUSION AND RECOMMENDATION

9.1 Conclusion

The following conclusions can be drawn from this study:

- There is a radial gas composition profile within the annulus of a spouted bed coal gasifier.
- The combustion reaction takes place within a very narrow region close to the gas inlet in the annulus and in the lower section of the bed in the spout.
- Pyrolysis in a spouted bed coal gasifier is not instantaneous. This process and the volatiles reforming reactions should be accounted for in order to construct the gas composition profiles both in the annulus and in the spout for the Forestburg coal-air system. There is a substantial amount of pyrolysis, especially in the upper section of the bed.
- No significant effects of the total bed height, char recycling and the average operating temperature (within range investigated) are observed on the measured gas composition profiles in the bed for the Forestburg coal-air system.
- The contribution from pyrolysis on the gas composition pattern in the bed for Forestburg coal-oxygen and Balmer coal-oxygen appears to be insignificant. However the effects of the volatiles reforming reactions as the source of carbon monoxide and hydrogen could be important.

- There is a large axial temperature variation in the spout to give the hottest spot in the upper section. For the Forestburg coal-air system operated under a relatively deep bed, the temperature profile along the centre of the annulus resembles those in a moving bed gasifier.
- The H₂/CO ratio along the bed of a spouted bed gasifier is lower when partially devolatilised char instead of coal is used as the feed.
- Both the one-dimensional model and the streamtube model give a very similar prediction in terms of the overall exit gas composition as well as the gas composition profiles along the centre of the annulus, but they could not be matched with the experimental results.
- The streamtube model is superior to the one-dimensional model as it is able to predict radial gas composition profiles over the entire annulus which have been observed experimentally.
- It is necessary to include the particle movement within the bed in order to accurately predict the performance of a spouted bed coal gasifier.

9.2 Recommendation

The following should be included for future work:

- Further study the effect of char recycle, bed height and temperature on the gas composition profiles for the Forestburg coal-oxygen and Balmer coal-oxygen system.

- Further confirm those profiles measured along the spout. In this case it is necessary to construct the actual spout region within the bed by taking radial pressure drop profiles at different bed levels.
- Investigate whether the water gas shift reaction is in equilibrium throughout the bed. For this purpose the steam composition profiles should also be determined.
- Perform more experiments on char gasification.
- Improve the gasification model. In this case the particle movement in the bed should be included as well as some form of radial dispersion in the annulus.

NOMENCLATURE

A _a	cross-sectional area of annulus, m ²
A _c	cross-sectional area of column, m^2
^A p	surface area of a single particle, m^2
A _s	cross-sectional area of spout, m ²
C _{ai}	concentration of component i in annulus gas, mol/m^3
C aiH	C_{ai} at $z = H$, mol/m ³
C _{azi}	concentration of component i along streamline in steamtube model, mol/m^3
C _{aziH}	C_{azi} at $z = H$, mol/m ³
C _{ei}	concentration of component i leaving reactor, mol/m^3
Cg	gas concentration, mol/cm^3 unless otherwise specified
C _s	carbon concentration of char; C_{so} , same at zero conversion, mol/cm ³ unless otherwise specified
C _{si}	concentration of component i in spout gas, mol/m^3
C _{siH}	C_{si} at $z = H$, mol/m^3
Cp	specific heat capacity, J/kg:K
D	diffusion coefficient; D', same for oxygen, ${ m cm}^2/{ m s}$
De	effective diffusivity in solid; D _{eo} , same at zero conversion, cm ² /s
d	column diameter, m
di	gas inlet diameter, m
^d p	particle diameter, m unless otherwise specified
d _s	spout diameter, m

Е'	activation energy for pyrolysis, kJ/mol
Е	activation energy for char-gas reaction, kJ/mol
F w	a parameter used in Equation 2.34
f ₁ , f _o	relative reactivity factor for $char-H_20$ reaction used for Johnson kinetics (Eqn. 2.22)
G	gas mass flow rate per unit of column cross-section, kg/m ² .s
Ga	Galileo number
g	acceleration of gravity (9.81 m/s^2)
H	bed depth, unexpanded, m
Hm	maximum spoutable bed depth, m
h	heat transfer coefficient, W/m ² .°K
К	equilibrium constant for char-gas reaction
K _{ws}	equilibrium constant for water gas shift reaction
k', k'	pyrolysis rate constant, s ⁻¹
k ₁ , k ₂ , k ₃	constants of Langmuir-Hinshelwood type rate expressions for char-CO $_2$ and char-H $_2$ O reaction
^k I	rate constant for char-H ₂ O reaction in Johnson kinetic (Eqn. 2.22)
k ash	ash film diffusion rate constant, g/cm ² .atm.s
k _c	thermal conductivity of coal, W/m.°K m
^k diff	diffusional reaction rate constant for char-gas reaction, g/cm^2 .atm.s
^k overall	overall rate constant for char-gas reaction as defined by Equation 6.7, g/cm^2 .atm.s
k _s , k _{so}	surface reaction rate constant for char-gas reaction g/cm ² • atm • s

.

k _v , k _{vo}	volumetric rate constant for char-gas reaction $(cm^3/mol)^n \cdot s^{-1}$
L	latent heat of vapourisation of water, J/kg
MC	coal moisture content, fraction, dry basis; MC _O , same at zero time
M _w	molecular weight of carbon, g/mol
^m p	mass of a single particle, kg/m^3
Nu	Nusselt number
n	order of reaction with respect to gaseous reactant concentration or partial pressure
P	total pressure, atm
P ₁ ,P ₂ , P ₃	Reactants partial pressure in Langumuir type rate equation, atm
Pg	gas partial pressure, atm
PL	path length along streamline, m
Pr	Prandalt number
Q _o	inlet gas flowrate, kg/h
Q(J-1)	volumetric gas flow along streamline J, m^3/s
R	universal gas constant (8.314 J/mol.°K)
R'	universal gas constant (82.06 atm.cm ³ /mol.°K)
R _c	column radius, m
Re	Reynolds number
(Re) bf	Reynolds number at beginning of fluidization
(Re) _{tf}	Reynolds number at complete fluidization
Rj	rate of jth reaction given by equation $6.1 - 6.3$, kmol/m ³ of particle volume.s

r _c	core radius at time t in the core shrinking model; r ,
	same at $t = 0$, m unless otherwise specified
r	radial distance from the column axis, m unless otherwise specified
Sc	Schmidt number
Т	temperature, °K
To	maximum temperature to which char has been exposed prior to gasification, °K
T _{wb}	adiabatic wet bulb temperature, °K
t	time, s
U a	superficial gas velocity in annulus, m/s
U _{aH}	U_a at z=H, m/s
U _{az}	superficial gas velocity along streamline, m/s
U _{bf}	superficial gas velocity at beginning of fluidization m/s
U _{mf}	minimum fluidization velocity, m/s
U ms	minimum spouting velocity, m/s
Ů _r	radial gas cross flow velocity at spout-annulus interface, m/s
U _s	gas spouting velocity, m/s
Ū _s	superficial gas velocity in spout, m/s
^U tf	superficial velocity at complete fluidization, m/s
^u s	gas sampling velocity; u _{iso} , same at isokinetic condition, m/s
V	volatiles loss from particle up to time t, fraction of original coal weight

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X vm	proximate volatile matter content of coal, gm/gm of coal (daf)
x	fraction of carbon conversion of char due to char-gas reaction
Y	r _c /r _o
2	vertical distance from gas inlet, m unless otherwise specified
Greek Symbols	
α	a parameter in Johnson kinetic, equation 2.22
a _v	relative pore surface area function in volumetric reaction model
3	voidage of the ash layer
ε a	annulus voidage
^e az	voidage along streamline
εs	spout voidage
^р Ъ	solid bulk density, kg/m^3 except for calculating D_s (Table 9) where it should be expressed in g/cm^3
ρ _c	molar density of carbon in coal particle, mol/cm^3
¢f	gas density, kg/m ³
^р р	particle density, kg/m^3 except for calculating H_m (Table 9) where it should be expressed in g/cm^3
ρ _{wc}	raw coal density, kg/m ³
η	effectiveness factor
ψ	product distribution of char combustion defined by Equation 2.17
λ	shape factor of particle. $\lambda = 1$ for spherical particle

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APPENDIX A

Estimation of Isokinetic Sampling Flowrate

The gaseous environment varies with respect to its temperature and composition from different points in the bed and from one run to the other. In order to estimate the sampling rate a gas temperature of 900°C and a gas mixture of 10% steam/90% air (v/v) has been assumed. These values were chosen based on the typical results obtained in some of the previous runs. Below is a sample calculation for point 6A.

Height above gas inlet, z (figure 7)	= 0.65 m
Coal particle diameter	= 0.0021 m
Column diameter	= 0.305 m
Gas inlet diameter	= 0.0318 m
Coal particle density	$= 1.3 \text{ Mg/m}^3$
Maximum spoutable bed height, H _m (Table 10)	= 2.03 m
Minimum fluidization velocity, U _{mf} (Table 10)	= 0.89 m/s
Superficial gas velocity, U _a (Table 10)	= 0.55 m/s

Interstitial gas velocity $\frac{U_a}{\varepsilon_a} = 1.29 \text{ m/s}$

Sampling probe cross-sectional area (1/4" dia. tube)

Isokinetic sampling volume at 900°C = $4.1 \times 10^{-5} \text{ m}^3/\text{s}$ Assuming that the gas leaving the water separator is saturated, the gas sampling volume at 21°C to be recorded by the rotameter is $1 \times 10^{-5} \text{ m}^3/\text{s}$ or 1.27 ft³/hr.

 $= 3.17 \times 10^{-5} m^2$

APPENDIX B

Operating Conditions and Experimental Results

Table 14

Operating conditions I

Run No.	Coal Type	H (cm)	Wet Coal Feed kg/h	Char Recycle kg/h	Cyclone Catch kg/h
88	Ł	72	40.20	0	N/A
89	F	. 38	44.40	0	N/A
90	F	22	43.80	12.96	4.97
91	F	30	50.40	3.57	7.25
93	F	97	55.80	3.57	8.51
94	F	23	47.58	0	6.40
96	F	35	48.00	0	7.97
103	В	1.28	37.20	0	16.29
106	FC	41	33.42	0	N/A

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F = Forestburg

B = Balmer

FC = Partially devolitilised Forestburg Char

N/A = not available

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operating conditions i	ET		s	11	n	o	i	t	1	1	d	n	0	C	g	n	i	t	a	r	e	p	0
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		Operating condition	ons II		
Run No.	Steam Flowrate kg/h	Air Flowrate kg/h	Pure O ₂ Flowrate kg/h	Pure N ₂ Flowrate kg/h	_
88	9.53	92.64	0	0	
89	10.53	113.23	0	0	
90	10.38	103,39	0	0	
91	11.95	100.61	0	0	
93	13.23	100.17	0	0	
94	119.28	0	31.47	0	
96	83.38	0	25.43	9.41	
103	72.55	0	26.41	0	
106	15.84	100.17	0	0	

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Table	16
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Operating conditions III

Run No.	H ₂ O/air (w/w)	H ₂ 0/0 ₂ (w/w)	H ₂ O/dry Coal (w/w)	Air/dry Coal (w/w)	0 ₂ /dry Coal (w/w)
88	0.103		0.31	3.02	- -
89	0.093	-	0.31	3.34	-
90	0.100	-	0.31	3.09	-
91	0.119	-	0.31	2.62	-
93	0.132	-	0.31	2.35	-
94	-	3.79	3.29	~	0.87
96	-	3.27	2.28	-	0.70
103	-	2.87	1.98		0.72
106	0.158	_	0.48	3.05	-

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Experimental results: Run 88

Point No	Тетр		Gas Composition, vol. % (dry)						
FOINT NO.	°C	°C	H ₂	CO ₂	02	N ₂	CH4	CO	
6A		_	_						
6B	-	-	11.232	15.436	0.180	63.444	1.673	8.045	
6C	-	-	8.510	10.844	1.312	67.421	0.874	11.039	
5A -	-	-	31.439	18.190	0.344	26.014	6.080	17.934	
5B	-	-	15.631	15.003	0.187	57.022	2.002	10.162	
5C	-	-	10.567	14.215	0	65.054	1.227	8.937	
4A	-	-	21.354	14.703	0.134	44.430	2.872	16.505	
4B	-		15.696	14.037	0.199	55.739	2.037	12.292	
4C	-	-	8.897	13.984	0.189	68.097	0.930	7.903	
3A	-	-	22.292	8.318	0.253	49.396	2.248	17.493	
3B	-		13.556	12.281	0.149	60.691	1.253	12.073	
3C	-	-	5.179	13.369	0.237	75.353	0.418	5.445	
2A		-	15.108	3.356	1.997	64.795	0.997	13.747	
2B	-		8.434	7.974	0.279	74.099	0.415	8.799	
2C	-	-	0.850	13.063	4.791	80.159	0	1.137	
1A	-	-	8.709	3.837	0.164	75.183	0.197	11.910	
1 B	-	-	- '	-		-		-	
1C	-	_ `	1.309	14.927	1.301	80.050	0	2.417	

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Experimental results: Run 89

Daint No	Tomo	Auto Tomo	Gas Composition, vol. % (dry)					*******
FOINE NO.	°C	°C	H ₂	co2	02	N ₂	CH4	CO
6A	-	.	18.050	8.002	0.161	50.847	1.537	21.403
6B	_	-	13.253	10.709	0.322	58.703	1.346	15.667
6C	-	-	11.016	12.862	0.050	63.412	1.264	11.394
5A		-	21.672	7.845	0.138	45.862	1.997	22.487
5B	-	-	13.878	10.212	0.110	57.723	1.400	16.676
5C		-	8.421	13.222	0.127	67.451	0.879	9.900
4A			30.307	8.872	0.109	28.440	3.428	28.844
4B	-		14.653	9.585	0	58.336	1.374	16.052
4C	_	-	3.897	14.330	0.115	75.656	0.570	5.432
3A	-	-	32.047	7.652	0.070	27.242	4.432	28.551
3B		-	17.613	8.262	0.230	55.846	1.863	16.187
3C	_	-	2.941	12.798	· 3.712	77.130	0.279	3.141
2A	-	-	27.463	3.141	0.090	45.740	2.727	20.831
2 B		-	12.264	5.960	0.772	68.251	1.039	11.714
2C	-	-	0.599	6.730	10.823	81.105	0	0.744
1A	-	-	11.967	1.547	0.207	67.527	0.424	18.328
1B	_	-	-			-	-	-
1C			0.206	13.175	6.217	80.402	0.	0

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Experimental results: Run 90

Daint No	Tomo t	* Aug Tomp	Gas Composition, vol. % (dry)					
POINT NO.	°C	°C	н ₂	co ₂	02	N ₂	CH4	CO
6A	899		16.491	9.528	0.088	50.479	1.317	21.648
6B	937	-	12.361	11.059	0.115	59.138	1.084	16.243
6C	973	-	8.531	12.374	0.162	67.831	0.748	10.354
5A	893	-	22.048	8.406	0.057	42.114	1.727	25.648
5B	933	-	10.883	12.687	0.967	60.394	0.999	14.070
5C	959	-	10.404	12.520	0.205	56.525	0.910	9.975
4A	761	-	22.420	8.974	0.142	51.859	3.511	13.094
4B	805	-	11.826	14.304	0.127	62.392	1.632	9.720
4C	840	-	5.728	12.817	1.458	73.426	0.713	5.859
3A	801	-	27.929	10.886	0.130	33.089	3.497	24.469
3B	841	. –	14.519	11.579	0.080	57.162	1.502	15.218
3C	884	-	3.334	10.484	4.869	76.088	0.295	4.931
2A	830	-	24.391	3.350	0.100	48.557	1.888	21.715
2B	859		6.242	10.017	0.109	74.135	0.310	9.179
2C	820	-	1.162	9.540	8.523	78.878	0.054	1.842
1A	837	_	8.225	3.837	0.148	72.010	0.186	15.594
1 B	855	-	5.140	8.299	2.449	73.948	0.147	10.016
10	766	-	1.089	11.662	7.239	77.719	0	2.291

*Recorded while sucking sample at any partcular point.

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Experimental results: Run 91

Gas Composition, vol. % (dry)									
Point No.	°C	Ave. Temp °C	H ₂	CO2	02	N ₂	СН4	CO	
6A	785	-	25.041	11.711	3.455	37.795	3.540	18.458	
6B	792	<u> </u>	22.086	13.044	1.595	42.295	3.077	17.904	
6C	805	-	12.000	14.960	4.283	56.065	1.601	11.089	
5A	764	-	27.718	19.666	0.312	30.418	6.501	15.322	
5B	· 780		20.604	17.744	0.131	43.436	4.429	13.655	
5C	795	-	11.170	13.987	1.461	60.434	2.332	10.615	
4A	-	_	23.278	14.013	0.541	25.610	3.169	19.414	- 1
4B	· -	-	17.599	14.341	0.150	49.297	2.231	16.382	1 L
4C		-	9.384	12.370	2.086	66.566	1.021	8.565	4 4
3A	764	-	25.722	15.087	0.492	36.478	3.946	18.275	1
3B	781	-	15.066	14.728	0.147	56.330	1.879	11.850	
3C	7 9 0	-	3.558	7.000	10.170	76.204	0.334	2.733	
2A	780	-	20.147	6.645	0.367	52.587	1.666	18.589	
2B	813	-	4.357	13.090	0.146	77.872	0.266	4.274	
2C	702	-	0.785	8.699	9.735	79.834	0	0.947	
1A	735	-	2.610	2.771	14.727	76.450	0.111	2.930	
1 B	740	-	4.307	12.416	0.126	77.166	0.129	5.850	
1C	680	-	0.126	8.969	9.500	80.798	0	0.607	

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Experimental results: Run 93

Point No	Temp *	Ave Temp **	Gas Composition, vol. % (dry)					
	°C	°C	Н ₂ .	CO2	02	N ₂	CH ₄	CO
6A	798	775	24.407	13.643	0.246	35.624	3.222	28.858
6B	803	782	19.623	13.722	0.205	41.684	2.571	19.118
6C	807	790	11.380	13.545	0.300	59.892	0.732	13.052
5A	726	776	22.833	14.006	0	36.348	4.819	22.000
5B	741	782	18.093	14.300	0.009	51.005	3.633	12.960
5C	753	792	12.037	13.180	0.300	61.759	1.674	11.05
4A	794	775	21.373	12.380	0	42.327	2.663	21.257
4B	807	784	15.206	12.970	0	54.787	1.649	15.388
4C	825	801	10.426	12.979	0.300	59.847	0.956	10.082
3A	746	773	22.465	14.584	0.326	41.555	3.389	17.681
3B	764	790	12.767	13.984	0.290	61.488	1.498	9.973
3C	755	797	3.964	12.098	1.811	78.270	0.442	3.415
2A	779	767	18.978	6.368	0.315	37.100	1.373	18.340
2B	809	803	6.676	11.516	0	75.016	0	6.792
2C	687	706	0.186	6.049	14.019	79.746	0	0
1A	751	763	8.051	8.756	0	74.334	0.232	8.627
1B	775	785	4.541	13.343	0	76.357	0.134	5.631
1C	659	680	0.210	8.840	10.784	79.248	0	0.919

*Recorded while sucking sample at any particular point.

**Will give temperature profile along radial positions A, B, C. Averaged over the run.

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Experimental results: Run 94

Point No	Tomp	Ave Tomo *	Gas Composition, vol. % (dry)					
°C	°C	°C	H ₂	C0 ₂	02	N ₂	СН4	СО
6A		867	-	_	_	-	-	-
6B	÷	887	-	-	-			
6C	-	881	-	-	-			
5A	_	852	-	-			-	-
5B	-	880	-	-	-		-	-
5C	-	894		-	·	-		
4A		830	-	-			-	
4B	- '	862	-	- .	-	-	-	
4C		870		-	-	- ,	-	
3A	-	816	-		-	-	-	-
3B	-	855	-			-	-	
30	-	842	-		-	-	-	
2A	-	804	-	-	-	-	-	-
2B	-	844	 .	-		-	-	-
2C	-	762		· –	-	-		<u> </u>
1A	- .	785	-	-	-	-	-	-
1 B	-	823	-	-	-	-	-	-
1C	-	720	-	-	-	-	-	<u> </u>

**Will give temperature profile along radial positions A, B, C. Averaged over the run.

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Experimental results: Run 96

Deint No	Tomo *	Ave Tomp	Gas Composition, vol. % (dry)					
FOINC NO.	°C	°C	H ₂	co ₂	02	N ₂	Сн ₄	CO
6A	785		37.201	28.872	3.007	11.333	0	19.586
6B	793	-	39.694	30.646	1.316	5.933	0	22.411
6C	805	-	29.290	36.655	1.368	15.069	0	17.618
5A	-	-	41.832	26.732	0.815	10.609	0	20.012
· 5B	-	-	44.725	30.295	0.523	5.050	0	19.407
5C	-	-	-		-	-	-	
4A	743	-	47.838	27.371	0.766	1.200	0	22.826
4B	751		46.611	28.550	0.492	1.571	0	22.776
4C	770	-	32.602	22.772	1.666	18.981	0	23.979
3A	820	-	49.588	20.924	0.410	1.140	0	27.939
3B	835	-	45.222	27.340	1.408	4.940	0	21.089
3C	918	-	24.989	42.7.54	3.017	13.964	0	14.275
2A	810	-	50.418	22.604	0.522	1.485	0	24.971
2B	853	-	43.010	29.516	0.495	7.247	0	19.732
2C	867	-	45.325	24.876	1.530	12.890	0	15.379
1A	754	-	46.430	32.745	0.180	0.584	0	20.061
1 B	765	-	43.562	33.948	0.783	2.225	0	19.483
10	815	-	21.966	46.103	3.316	15.296	0	12.319

*Recorded while sucking sample at any particular point.

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Experimental results: Run 103

	Temp. °C	Ave. Temp. °C	Gas Composition, vol. % (dry)						-
Point No.			H ₂	co ₂	02	N ₂	CH4	CO	—
6A		_ _	43.831	28.471	0.573	0	1.405	25.720	
6B	-	-	40.670	26.377	1.212	0	1.532	30.209	
6C	-	_		-	_		-	-	
5A	-	-	42.636	28.214	0.300	0	2.444	26.406	
5B			39.142	29.246	0.504	0	1.636	29.476	I.
5C								-	Ŀ
4A	_	-	40.615	30.206	1.130	0	2.049	26.000	68
4B	-	-	36.270	29.645	3.61	0	1.538	28.917	i
4C		-		-	-	-	-	-	
3A	-	-	39.032	33.590	0.080	0	1.633	25.635	
3B		-	34.528	34.758	2.968	0	1.839	29.907	
3C	-	-	-	- `·	-	-	. -	-	
2A		-	37.925	36.012	0.133	0	1.107	24.823	
2B	_	-	29.822	43.822	0.096	0	0.957	25.303	
2C	_	-	-	_	_		-	_	
1A		-	-		-	-	-	-	
1 B	_	_		_	_	-	· _		
1C			-					-	

Table 25

Experimental results: Run 106

Defet Ne	Temp. °C	Ave. Temp. °C	Gas Composition, vol. % (dry)					
Point No.			H ₂	co ₂	02	N ₂	СН4	CO
6A	981	904	8.020	13.471	0.355	66.656	0.265	11.232
6B	885	931	11.692	12.890	0.951	59.667	0.460	14.340
6C	9 08	921	7.094	9.521	6 .9 00	69.195	0	7.290
5A	943	896	12.315	13.666	0.070	59.612	0.440	13.890
5B	867	917	13.878	9.852	1.501	54.350	0.616	19.803
5C	897.	922	10.593	9.048	5.391	61.868	0.350	12.651
4A	946	886	14.538	9.806	1.341	56.420	0.417	17.478
4B	857	898	14.497	8.808	2.012	50.575	0.817	23.291
4C	902	918	9.010	6.130	5.201	68.094	0.354	11.211
3A	922	880	24.692	6.438	0.028	36.612	0.906	31.325
3B	. 874	881	11.916	6.435	5.642	60.671	0.500	14.838
3C	904	900	7.253	10,986	2.739	71.229	0.226	7.568
2A	913	873	27.652	2.067	1.196	36.284	0.764	32.037
2B	855	879	8.264	9.807	3.720	69.989	0.231	7.989
2C	913	850	4.802	12.506	2.264	75.604	0	4.821
1A		-	-		-	-	-	-
1 B	-	-		-	-	-	-	-
1C	-	-		-	-	-	-	_

*Recorded while sucking sample at any particular point.

**Will give temperature profile along radial positios A, B, C. Averaged over the run.

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I.

Experimental results: exit gas composition and average bed temperature

Dum No	Accorden Ded Tomo	Gas Composition, vol. % (dry)						
Kun No.	°C	H ₂	CO ₂	02	N ₂	CH4	CO	
88	776	13.601	14.971	0	58.136	2.020	11.271	
89	877	12.780	12.660	0	58.020	1.290	15.250	
90	907	12.800	12.440	0	58.210	1.120	15.430	I
91	786	14.431	14.611	0	56.586	1.810	12.561	170 -
93	800	14.190	16.350	0	57.460	2.140	9.860	1
94	960	35.266	41.376	0	0.640	2.600	20.118	
96	788	36.850	34.200	0	8.09	0	20.860	
103	918	37.759	29.918	0	0	3.492	28.831	
106	901	11.011	14.231	0	61.686	0.300	12.771	

APPENDIX C

Gas Chromatograph Calibration Table

CAL	RT	LEVEL	AMT	AMTZAREA	REF	NAME
1	3.697	1	2.04800E+01	8.23932E-04		H2
		2	1.63800E+01	8.12137E-04		
		3	1.22900E+01	8.01653E-04		
		4	8.19000E+00	7.72530E-04		
		5	4.10000E+00	7.68445E-04		
2	5.421	1	2.01100E+01	1.02101E-02		C02
		2	1.60900E+01	9.93132E-03		
		3	1.20700E+01	9.90794E-03		
		4	8.04000E+00	9.67687E-03		
		5	4.02000E+00	8.89499E-03		
3	7.852	1	1.00000E-07	2.24140E-09		02
		2	4.20000E+00	7.19356E-03		
		3	8.40000E+00	6.99032E-03		
		4	1.26000E+01	6.99249E-03		
		5	1.63000E+01	6.95213E-03		
4	9.198	1	3.26900E+01	8.52444E-03	+	N2
		2	4.19600E+01	8.96309E-03		
		3	5.12100E+01	9.06236E-03		
		4	6.04800E+01	9.19693E-03		
		5	6.97300E+01	9.22045E-03		
5	12.314	1	4.08000E+00	5.03819E-03		CH4
		2	3.26000E+00	4.61051E-03		
		3	2.45000E+00	5.11641E-03		
		4	1.63000E+00	5.42003E-03		
		5	8.16000E-01	5.71754E-03		
6	16.253	1	2.26400E+01	9.67058E-03		C0
		2	1.81100E+01	9.50005E-03		4
		3	1.35800E+01	9.62132E-03		
		4	9.06000E+00	9.96048E-03		•
		5	4.53000E+00	9.67799E-03		

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APPENDIX D

Computer Simulation

Table 27

Bed geometry and properties of solids used in model simulation

Parameters	Values
Column ID	0.305 m
Cone angle (included)	60 °
Height of conical section	19.8 cm
Orifice diameter	3.18 cm
Particle density	1.3 g/cm ³
Solid bulk density	0.523 g/cm^3
Annulus voidage	0.43
Particle diameter (spherical)	2.1 mm

Computer Programme for the One-dimensional Model

C This programme simulates the axial gas composition C profiles and the exit gas composition. C Water gas shift equilibrium is assumed. Bed is C assumed to be isothermal. The fourth order Runge-C Kutta technique from a library subroutine has C been used to solve the differential equations. Gas distribution due to pyrolysis has been С C theoretically predicted. This model is based on C the gas plug flow in annulus and spout. С сн -Total bed height C TEMP -Bed temperature C P -Operating pressure -Frequency factor for water gas shift reaction C RWSE C EWSE -Activation energy for water gas shift reaction -Frequency factor for C/H20 reaction RSC С -Activation energy for C/H2O reaction C ESC -Frequency factor for C/CO2 reaction C RCDC -Activation energy for C/CO2 reaction -Frequency factor for C/O2 reaction C ECDC С ROC C EOC -Activation energy for C/02 reaction C DC -Column diameter(m) C DP -Particle diameter(m) С DI -Orifice inlet diameter(m) -Coal density(g/m3) С RAWC С RAWB -Bulk density of coal(g/m3) -Overall gas flowrate(kg/h) C TGF -Number of DE's to be solved С Ν -Initial bed height С X С Y(1) -Initial H20 conc. in spout(mol/m3) С -Initial H20 conc. in annulus(mol/m3) Y(2) -Initial CO conc. in spout(mol/m3) С Y(3) С Y(4) -Initial CO conc. in annulus(mol/m3) -Initial H2 conc. in spout(mol/m3) C Y(5) С Y(6) -Initial H2 conc. in annulus(mol/m3) -Initial 02 conc. in spout(mpo1/m3) С Y(7) C Y(8) -Initial 02 conc. in annulus(mol/m3) -Initial CO2 conc. in spout(mol/m3) C Y.(9) C Y(10) -Initial CO2 conc. in annulus(mol/m3) -Initial N2 Conc. in spout & annulus(mol/m3) C CN2 -A factor in Mammuro-Hattori equation C CUMF СК -Number of steps in the integration C EA -Bed porosity in annulus -Gas viscosity(Ns/m2) C VIS IMPLICIT REAL*8(A-H,O-Z) DIMENSION Y(10), F(10), T(10), S(10), G(10) DIMENSION HA(27), HS(27), CMS(27), CMA(27), OA(27), OS(27). #CDA(27), CDS(27), HEI(27), H2OA(27) COMMON/BLKA/UMFH, RAWGH, DP, CUMF, TEMP COMMON/BLKB/H, CN2, P, RWSE, EWSE, RSC, ESC, PIE, DC. #RCDC, ECDC, ROC, EOC, AS, HM, RAWC, GMF, K COMMON/BLKC/I EXTERNAL FUNC DATA N.X/10..003D0/ DATA Y/1.907D0,1.907D0,0.D0,0.D0,0.D0,0.D0,1.998D0,1.998D0, #0.000D0,.000D0/ K=27 H=0.97D0 TEMP=1073.DO CN2=7.515D0

```
P=1.D0
     RWSE = .0265D0
     EWSE=3955.DO
     RSC=1.2D9
     ESC=29790.DO
     RCDC=4.0D8
     ECDC=29790.DO
     RDC=860.DO
     E0C=18000.D0
     DC=.305D0
     RAWC=1.3D6
     DP=.00210D0
     DI=.0318D0
     TGF=118.0D0
     RAWB = . 523D6
     CUMF = 0.9D0
     PIE=4.DO*DATAN(1.DO)
     ACH=PIE*DC*DC/4.DO
C Maximum Spoutable bed height HM
     HM=.105DO*DC*((DC/DP)**.75DO)*((DC/DI)**.4DO)/
    #((RAWC/1.D6)**1.2D0)
С
C Mean spout diameter
     GMF = (TGF)/3600.D0
     DS=.118DO*((GMF/ACH)**.49DO)*(DC**.68DO)/
    #((RAWB/1.D6)**.41DO)
     AS=PIE*DS*DS/4.DO
     AAH=ACH-AS
С
C Calculate H1, HMIN, set E required for DRKC
     H1=(H-X)/64.DO
     HMIN=0.01DO*H1
     E=1.D-5
C Call DRKC to solve DE's
     DELZ=(H-X)/ FLOAT(K)
     Z=X+DELZ
     WRITE(6,101)
     FORMAT(/'PERCENTAGE DRY GAS COMPOSITION IN SPOUT'/)
101
     DO 40 I=1.K
     CALL DRKC(N,X,Z,Y,F,H1,HMIN,E,FUNC,G,S,T)
С
C Concentration adjustment due to water shift reaction
     EWS=RWSE*DEXP(EWSE/TEMP)
     DELCS=(EWS*Y(1)*Y(3)-Y(5)*Y(9))/(Y(5)+Y(9)+EWS*(Y(1)+Y(3)))
     DELCA=(EWS*Y(2)*Y(4)-Y(6)*Y(10))/(Y(6)+Y(10)+EWS*(Y(2)+Y(4)))
     Y(1)=Y(1)-DELCS
     Y(2)=Y(2)-DELCA
     Y(3)=Y(3)-DELCS
     Y(4) = Y(4) - DELCA
     Y(5)=Y(5)+DELCS
     Y(6)=Y(6)+DELCA
     Y(9)=Y(9)+DELCS
     Y(10)=Y(10)+DELCA
     IF(Y(7), LT.O.)Y(7)=O.DO
     IF(Y(8).LT.O.)Y(8)=0.DO
     Z=X+DELZ
     H2OA(I)=Y(2)
C Calculate percentage dry gas composition at each level
     TMDA=CN2+Y(4)+Y(6)+Y(8)+Y(10)
```

```
CMS(I)=Y(3)/TMDS*100.DO
     CMA(I)=Y(4)/TMDA*100.DO
     HS(I)=Y(5)/TMDS*100.DO
     HA(I)=Y(G)/TMDA*100.DO
     OS(I) = Y(7) / TMDS * 100.DO
     OA(I)=Y(8)/TMDA*100.DO
     CDS(I)=Y(9)/TMDS*100.DO
     CDA(I) = Y(10) / TMDA * 100.D0
     HEI(I)=X
     WRITE(6,21)HEI(I),CMS(I),HS(I),CDS(I),OS(I)
21
     FORMAT(2X, 5F10.3)
     CONTINUE
40
С
     UAH=CUMF*UMFH*(1.DO-((1.DO-H/HM)**3))
     USH=(GMF-(UAH*AAH*RAWGH))/(AS*RAWGH)
C Calculation of percentage dry gas composition (exit)
     CH20E=((UAH*AAH*Y(2))+(USH*AS*Y(1))+.07388D0)
     CCDE = ((UAH * AAH * Y(4)) + (USH * AS * Y(3)) + .04084D0)
     CH2E=((UAH*AAH*Y(6))+(USH*AS*Y(5))+.1266DO)
     CO2E=((UAH*AAH*Y(8))+(USH*AS*Y(7)))
     CCO2E=((UAH*AAH*Y(10))+(USH*AS*Y(9))+.01377D0)
     CH4E=.011050
     CN2E=((UAH*AAH*CN2)+(USH*AS*CN2))
C Changes in concentration due to gas combustion and
C water shift equilibrium adjustment
     DE0=C02E/(CC0E+CH2E)
     CH20E1=CH20E+2.DO*DE0*CH2E
     CCDE1=CCDE-2.DO*DEO*CCDE
     CH2E1=CH2E-2.DO*DE0*CH2E
     CC02E1=CC02E+2.DO*DE0*CC0E
     C02E=0.D0
     IF(CCOE1.LE.O.)GO TO 222
     IF(CH2E1.LE.O.) GO TO 222
     DELCF=(EWS*CH20E1*CC0E1-CH2E1*CC02E1)/
    #(CH2E1+CCO2E1+EWS*(CH2OE1+CCOE1))
     CH2OF = CH2OE 1 - DELCF
     CCOF=CCOE1-DELCF
     CH2F=CH2E1+DELCF
     CC02F=CC02E1+DELCF
    TCED=CC0F+CH2F+C02E+CC02F+CN2E+CH4E
     CMD=CCOF/TCED*100.DO
     HD=CH2F/TCED*100.DO
     OD=C02E/TCED*100.D0
     CCD=CC02F/TCED*100.DO
     CH4F=CH4E/TCED*100.DO
     WRITE(6,56)
     FORMAT(/'EXIT GAS COMPOSITION(% v/v DRY)'/)
56
     WRITE(6,50)CMD
50
     FORMAT(2X, 'CARBON MONOXIDE=',2X,F10.3/)
     WRITE(6,55)HD
55
     FORMAT(2X, 'HYDROGEN=', 2X, F10.3/)
     WRITE(6,60)0D
     FORMAT(2X, 'OXYGEN=', 2X, F10.3/)
60
     WRITE(6,65)CCD
     FORMAT(2X, 'CORBON DIOXIDE=', 2X, F10.3/)
65
     WRITE(6,66)CH4F
     FORMAT(2X, 'METHANE=', 2X, F10.3/)
66
     GO TO 74
222
     WRITE(6,224)
```

TMDS=CN2+Y(3)+Y(5)+Y(7)+Y(9)

```
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```

```
FORMAT(/2X, 'ALL CO OR H2 CONSUMED BY COMBUSTION IN FOUNTAIN'/)
224
     WRITE(6,75)
74
     FORMAT(/12X, 'PERCENTAGE DRY GAS COMPOSITION IN ANNULUS'/)
75
     WRITE(6,80)
     FORMAT(2X, 'HEIG(M)', 6X, 'C MONOX', 7X, 'HYDROGEN',
80
    #9X,'C DIOX', 10X,'OXYGEN'/)
     WRITE(6,85)(HEI(I),CMA(I),HA(I),CDA(I),OA(I),I=1,K)
     FORMAT(5F12.3)
85
     WRITE(6,111)H, DP, TEMP, RSC, RCDC
    FORMAT(/2X,'H=',F6.3,2X,'DP=',F6.5,2X,'TEMP=',F10.3,
111
    #2X, 'RSC=', E10.3, 2X, 'RCCD=', E10.3)
     STOP
     END
С
C Subroutine FUNC
     SUBROUTINE FUNC(X,Y,F)
     IMPLICIT REAL*8(A-H.O-Z)
     DIMENSION Y(10), F(10)
     COMMON/BLKA/UMFH, RAWGH, DP, CUMF, TEMP
     COMMON/BLKB/H, CN2, P, RWSE, EWSE, RSC, ESC, PIE, DC.
    #RCDC, ECDC, ROC, EOC, AS, HM, RAWC, GMF, K
     COMMON/BLKC/I
С
C Calculate gas density in spout, annulus and UMF
     VIS=4.61D-5
     RAWG=P*1.D5*29.D-3/(8.314DO*TEMP)
     GAN=(DP**3)*((RAWC/1.D3)-RAWG)*RAWG*9.81DO/(VIS**2)
     REBF=(((18.1D0**2)+.0192D0*GAN)**.5)-18.1D0
     RETF=(((24.DO**2)+.0546DO*GAN)**.5)-24.DO
     UBF=REBF*VIS/(DP*RAWG)
     UTF=RETF*VIS/(DP*RAWG)
     UMF=.5DO*(UBF+UTF)
     IF (I.LT.K) GO TO 25
     UMFH=UMF
     RAWGH=RAWG
C Superficial velocity in annulus
25
      UA=CUMF*UMF*(1.DO-((1.DO-X/HM)**3))
С
C Porosity in spout
      ES=1.DO-.2DO*X/H
C
C Average superficial vel. in spout and derivative of
C annulus area divide by annulus area(DIVA)
     IF(X.GE.O. 198DO) GO TO 10
     CRAD=X*(DTAN(PIE/6.DO))+0.0400D0
     AC=PIE*CRAD*CRAD
     DA1=DTAN(PIE/G.DO)
     DAZ=2.DO*PIE*((X*DA1*DA1)+0.0400DO*DA1)
     GO TO 20
10
     AC=PIE*DC*DC/4.DO
     DAZ=O.
     US=(GMF-(UA*(AC-AS)*RAWG))/(AS*RAWG)
20
     DIVA=DAZ/(AC-AS)
С
C Annulus vel. gradient DUA
     DUA=3.0D0*CUMF*UMF*((1.D0-X/HM)**2)/HM
     IF(Y(7).LT.O.)Y(7)=0.DO
     IF(Y(8), LT, O, )Y(8)=0.DO
С
```

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```
C Total gas concentration in spout
     TCS=Y(1)+Y(3)+Y(5)+Y(7)+Y(9)+CN2
C Total gas concentration in annulus
     TCA=Y(2)+Y(4)+Y(6)+Y(8)+Y(10)+CN2
C Rate constant for C/H2O reaction
     R1=RSC*(DEXP(-ESC/TEMP))*6.DO/DP
С
C Rate constant for C/CO2 reaction
     R2=RCDC*(DEXP(-ECDC/TEMP))*6.DO/DP
С
C Rate constant for C/O2 reaction
     R3=ROC*(DEXP(-EOC/TEMP))*6.D5/(DP*12.D0)
С
С
С
 DE for steam in spout
     F(1) = -R1*Y(1)*(1.DO-ES)/US
С
C DE for steam in annulus
     F(2) = -(R1*Y(2))*.57DO/UA+(Y(1)-Y(2))*
    #(DUA/UA+DIVA)
С
C DE for C Monox in spout
     F(3)=(R1*Y(1)+2.DO*R2*Y(9))*(1.DO-ES)/US
С
C DE for C Monox in annulus
     F(4)=(R1*Y(2)+2.DO*R2*Y(10))*.57DO/UA+
    #(Y(3)-Y(4))*(DUA/UA+DIVA)
С
C DE for Hydrogen in spout
     F(5)=R1*Y(1)*(1.DO-ES)/US
С
C DE for Hydrogen in annulus
     F(6) = (R1 * Y(2)) * .57DO/UA + (Y(5) - Y(6)) * (DUA/UA + DIVA)
С
C DE for Oxygen in spout
     F(7)=-R3*Y(7)/TCS*(1.DO-ES)/US
С
C DE for Oxygen in annulus
     F(8) = -R3*Y(8)/TCA*.57DO/UA+(Y(7)-Y(8))*(DUA/UA+DIVA)
С
C DE for C Diox in spout
     F(9)=(R3*Y(7)/TCS-R2*Y(9))*(1.DO-ES)/US
С
C DE for C Diox in annulus
     F(10)=(R3*Y(8)/TCA-R2*Y(10))*.57DO/UA+
    #(Y(9)-Y(10))*(DUA/UA+DIVA)
     RETURN
     END
```

Computer Programme for the Streamtube Model

```
C This programme uses the streamtube model to
C simulate gasification process. Water gas shift
C equilibrium is assumed.Bed is assumed to be
C isothermal. The fourth order Runge-Kutta technique
C from a library subroutine has been used to solve
C the differential equation.
С
      IMPLICIT REAL*8(A-H,O-Z)
      COMMON/BLKA/TEMP, UMF, RAWG, CUMF, HM, AS, HC,
     #RB, DC, PIE, GMF, H
      COMMON/BLKB/UST.EA
      COMMON/BLKC/R1,R2,R3,CN2
      DIMENSION R(81,41),Q(40),HS(40),HH(40),Y(5)
      DIMENSION F(5), YY(5), FF(5), TH(81), UA1(81), RC(81, 41)
      DIMENSION H205(40), COS(40), H25(40), CO25(40), O25(40)
      DIMENSION T(5), S(5), G(5)
      EXTERNAL FUNC1, FUNC2
      DATA Y/1.9073D0.0.D0.0.D0.0.D0,1.9976D0/
С
сн
         -Total bed height
C TEMP
         -Bed temperature
CP
         -Operating pressure
C RWSE
         -Frequency factor for water gas shift reaction
C EWSE
         -Activation energy for water gas shift reaction
C RSC
         -Frequency factor for C/H20 reaction
         -Activation energy for C/H2O reaction
-Frequency factor for C/CD2 reaction
C. ESC
С
  RCDC
         -Activation energy for C/CD2 reaction
C ECDC
C ROC
         -Frequency factor for C/02 reaction
  EOC
         -Activation energy for C/02 reaction
С
C DC
         -Column diameter(m)
C DP
         -Particle diameter(m)
C DI
         -Orifice inlet diameter(m)
. C
  RAWC
         -Coal density(g/m3)
C RAWB
         -Bulk density of coal(g/m3)
C TGF
         -Overall gass flowrate(kg/h)
С
  NN
         -Number of DE's to be solved
CN
         -Number of horizontal grid points
CM
         -Number of vertical grid points
С
  X
         -Initial bed heght(m)
С
  Y(1)
         -Initial H20 conc. in spout(mol/m3)
         -Initial CO conc. in spout(mol/m3)
С
  Y(2)
         -Initial H2 conc. in spout(mol/m3)
С
  Y(3)
         -Initial CO2 conc. in spout(mol/m3)
-Initial O2 conc. in spout(mol/m3)
С
  Y(4)
С
  Y(5)
         -A factor in Mammuro-Hattori eqn.
C CUMF
ск
         -Number of steps in the integration
C EA
         -Bed porosity in annulus
С
  VIS
         -Gas viscosity(Ns/m2)
С
  RB
         -Radial distance from centre of column to
С
          wall at the base of gasifier(m)
С
  CN2
          Initial N2 conc. in spout(mol/m3)
         -Height of cylindrical section(m)
С
  HC
C
  DZS
         -Incremental spout height to be specified
С
          required to calculate heights of streamlines
С
           in spout
Ċ
         -Tolerable error to be specified to calculate
  ER
С
          volumetric flow along streamlines(m3/s). These
c
          floerates are used to construct streamlines
С
          along the spout.
```

```
c zo
        -Spout height at inlet=0.
     H= .97DO
     CN2=7.515D0
     P=1.DO
     RWSE = . 0265D0
     EWSE=3955.DO
     RSC=3.D8
     ESC=29790.DO
     RCCD=1.D8
     ECCD=29790.D0
     ROC=860.D0
     EOC=18000.D0
     DC=.305D0
     RAWC=1.3D6
     DP=.0021D0
     DI=.0318D0
     TGF = 118.00D0
     RAWB= . 523D6
     CUMF = . 9DO
     EA=.43D0
     TEMP=1073.D0
     N=21
     M = 41
     NN=5
     RB=.0400D0
     HC=.198DO
     DZS=.010D0
     ER=1.D-5
     Z0=0.D0
С
     PIE=4.DO*DATAN(1.DO)
     ACH=PIE*DC*DC/4.00
С
C Mean spout diameter and max spoutable height
     GMF = (TGF)/3600.DO
     DS=.118DO*((GMF/ACH)**.49DO)*(DC**.68DO)/
    #((RAWB/1.D6)**.41DO)
     AS=PIE*DS*DS/4.DO
     HM=.105D0*DC*((DC/DP)**.75D0)*((DC/DI)**.4D0)/
    #((RAWC/1.D6)**1.2D0)
С
C Minimum fluidizing velocity and annulus velocity
C at top of bed
     VIS=4.D-5
     RAWG=P*1.D5*29.D-3/(8.314DO*TEMP)
     GAN=(DP**3)*((RAWC/1.D3)-RAWG)*RAWG*9.81DO/
    #(VIS**2)
     REBF=(((18.1D0**2)+.0192D0*GAN)**.5)-18.1D0
     RETF=((((24.DO**2)+.O546DO*GAN)**.5)-24.DO
     UBF=REBF*VIS/(DP*RAWG)
     UTF=RETF*VIS/(DP*RAWG)
     UMF = . 5DO*(UTF+UBF)
     UAH=CUMF*UMF*(1.DO-(1.DO-H/HM.)**3)
С
C Divide top of bed into N~1 sections and comput
C grid points along top of bed and column wall
С
     NM1=N-1
     MM1=M-1
     R(M, 1) = DC/2.DO
```

```
RI=(DC-DS)/(2.DO*NM1)
     DO 10 J=1,NM1
     R(M, J+1) = R(M, J) - RI
     CONTINUE
 10
     DZA=(H-ZO)/MM1
     Z=ZO
     DO 20 I=1,MM1
     IF(Z.LE.HC) R(I,1)=Z*(DTAN(PIE/6.DO))+RB
     IF(Z.GT.HC) R(I,1)=DC/2.DO
     Z=Z+DZA
 20
     CONTINUE
С
C Calculate volumetric flow from each streamtube
     DO 30 J=1,NM1
     Q(J) = PIE * (R(M, J) * *2 - R(M, J+1) * *2) * UAH * EA
 30
     CONTINUE
С
C Compute height of streamtube along spout
     Z1=0.D0
     QAO=0.DO
     DO 40 J=1.NM1
     JJ=0
 55
     Z=Z1+DZS
 60 UA=CUMF*UMF*(1.DO-(1.DO-Z/HM)**3)
     IF(Z.LE.HC) CRAD=Z*(DTAN(PIE/6.DO))+RB
IF(Z.GT.HC) CRAD=DC/2.DO
     AC=PIE*CRAD*CRAD
     QA=UA*(AC-AS)*EA
     DQA=QA-QAO
     E=DABS(Q(J)-DQA)
     IF(E.LE.ER) GO TO 45
     IF(JJ.EQ.1) GD TD 65
     IF(DQA.GT.Q(J)) GO TO 50
С
C Incremental search
     Z1=Z
     DQ1=DQA
     GO TO 55
С
C Linear interpolation
 50 JJ=1
     Z2=Z
     DQ2=DQA
     F1=DQ1-Q(J)
     F2=DQ2-Q(J)
     Z3=(Z1*F2-Z2*F1)/(F2-F1)
 75
     Z=23
     GO TO 60
65
     F3=DQA-Q(J)
     IF(F1*F3.LE.O.) GO TO 70
     Z1=Z3
     F1=F3
     GO TO 75
70
     Z2=Z3
     F2=F3
     GO TO 75
 45
     HS(J)=Z
     QAO=QA
     Z1=Z
С
```

```
C Compute mid point for each streamtube along spout
     IF(J.EQ.1) HH(J)=HS(J)/2.DO
     IF(J.GE.2) HH(J)=(HS(J)+HS(J-1))/2.DO
     CONTINUE
 40
С
C Rate constant for C/H20
     R1=RSC*DEXP(-ESC/TEMP)*6.DO/DP
C Rate constant for C/CD2
     R2=RCCD*DEXP(~ECCD/TEMP)*6.DO/DP
C Rate constant for C/02
     R3=RDC*DEXP(-EOC/TEMP)*6.D5/(12.DO*DP)
С
C Compute gas concentration frofiles at all inlet
C to streamtubes
C Calculate H1, HMIN, set ERC required by DRKC
     WRITE(6,77)
     FORMAT(' STREAMTUBE INLET CONDITIONS(DRY)'/)
 77
     WRITE(6,78)
 78
     FORMAT(2X, 'INLAT HT', 4X, 'CMONOX', 4X, 'HYD', 4X, 'CDIOX', 4X, 'OXY'/)
     ERC=1.D-5
     X=ZO
     DO 100 J=1,NM1
     H1=(HH(J)-X)/64.DO
     HMIN= .0100*H1
     ZZ = HH(J)
     CALL DRKC(NN,X,ZZ,Y,F,H1,HMIN,ERC,FUNC1,G,S,T)
     X=ZZ
C Concentration adjustment due to shift reaction
     WSE=RWSE*DEXP(EWSE/TEMP)
     DELC=(WSE*Y(1)*Y(2)-Y(3)*Y(4))/(Y(3)+Y(4)+WSE*(Y(1)+Y(2)))
     Y(1)=Y(1)-DELC
     Y(2)=Y(2)-DELC
     Y(3)=Y(3)+DELC
     Y(4) = Y(4) + DELC
     IF(Y(5).LE.O.) Y(5)=0.
     H20S(J)=Y(1)
     CDS(J)=Y(2)
     H2S(J)=Y(3)
     CO2S(J)=Y(4)
     02S(J)=Y(5)
С
C Calculate percent dry gas composition in spout

TMDG=CN2+Y(1)+Y(2)+Y(3)+Y(4)+Y(5)
     CMD=Y(2)/TMDG*100.DO
     HD=Y(3)/TMDG*100.DO
     CDD=Y(4)/TMDG*100.DO
     OD=Y(5)/TMDG*100.DO
     WRITE(6,90) HH(J),CMD,HD,CDD,OD
90
     FORMAT(5F12.3)
     CONTINUE
100
С
C Compute R(M,J)
     XX=O.DO
     DO 110 J=1,NM1
     YY(1) = H2OS(J)
     YY(2)=COS(J)
     YY(3)=H2S(J)
     YY(4)=CO2S(J)
     YY(5)=02S(J)
     K = 1
```

```
DO 120 I=2.M
     TH(I) = FLOAT(I-1) * DZA + ZO
     IF(TH(I).LT.HS(J)) GD TO 120
     IF(J.ÉQ.NM1.AND.I.EQ.M) K=1
     IF(I.EQ.M) GO TO 125
     UA1(I)=CUMF*UMF*(1.DO-(1.DO-TH(I)/HM)**3)
     W=(R(I,J)**2)-Q(J)/(UA1(I)*PIE*EA)
     R(I, J+1) = DSQRT(W)
С
C Calculate centre point at each level in streamtube
125 RC(I,J)=(R(I,J+1)+R(I,J))/2.DO
     IF(K.EQ.1) PL=DSQRT((TH(I)-HH(J))**2+(RC(I,J)-DS/2.DO)**2)
     IF(K.EQ.2) PL=DSQRT((TH(I)-TH(I-1))**2
    #+(RC(I,J)-RC(I-1,J))**2)
     IF(K.EQ.1) UAA=CUMF*UMF*(1.DO-(1.DO-HH(J)/HM)**3)
     IF(K.EQ.2) UAA=UA1(I-1)
     IF(K.EQ.1) A=TH(I)-HH(J)
     IF(K.EQ.2) A=TH(I)-TH(I-1)
     UST=UAA*A/PL
С
C Calculate gas concentration profiles in streamtube
     ZF=XX+PL
     H1=PL/64.DO
     HMIN=.01DO*H1
     CALL DRKC(NN, XX, ZF, YY, FF, H1, HMIN, ERC, FUNC2, G, S, T)
С
C Concentration adjustment due to water shift
     WSE=RWSE*DEXP(EWSE/TEMP)
     DELC=(WSE*YY(1)*YY(2)-YY(3)*YY(4))/(YY(3)+YY(4))
    #+WSE*(YY(1)+YY(2)))
     YY(1)=YY(1)-DELC
     YY(2)=YY(2)-DELC
     YY(3)=YY(3)+DELC
     YY(4) = YY(4) + DELC
     IF(YY(5).LT.O.) YY(5)=0.
С
C Calculate percent dry gas composition
TMDGA=CN2+YY(1)+YY(2)+YY(3)+YY(4)+YY(5)
     CMDA=YY(2)/TMDGA*100.DO
HDA=YY(3)/TMDGA*100.DO
     CDDA=YY(4)/TMDGA*100.DO
     ODA=YY(5)/TMDGA*100.DO
     K=2
     WRITE(6,140)TH(I),J,RC(I,J),I
140 FORMAT(2X, 'HEIG=', F10.4, 4X, 'J=', I2, 4X, 'DIS FR CENTRE=', F10.5,
    #4X,12)
     WRITE(6,145)CMDA,HDA,CDDA,ODA
     FORMAT(4F10.3/)
145
120
     CONTINUE
110
     CONTINUE
     STOP
     END
С
C Subroutine FUNC1
     SUBROUTINE FUNC1(X,Y,F)
     IMPLICIT REAL*8(A-H,O-Z)
     COMMON/BLKA/TEMP, UMF, RAWG, CUMF, HM, AS, HC, RB, DC,
    #PIE,GMF,H
     COMMON/BLKC/R1,R2,R3,CN2
     DIMENSION Y(5),F(5)
```

```
С
C Superficial velocity in annulus
    UA=CUMF*UMF*(1.DO-(1.DO-X/HM)**3)
С
C Porosity in spout
    ES=1.DO-.2DO*X/H
С
C Velocity in spout
     IF(X.LE.HC) CRAD=X*(DTAN(PIE/6.DO))+RB
     IF(X.GT.HC) CRAD=DC/2.DO
     AC=PIE*CRAD*CRAD
    US=(GMF-UA*(AC-AS)*RAWG)/(AS*RAWG)
    TCS=Y(1)+Y(2)+Y(3)+Y(4)+Y(5)+CN2
     F(1) = -R1 * Y(1) * (1.DO-ES)/US
     F(2)=(R1*Y(1)+2.DO*R2*Y(2))*(1.DO-ES)/US
     F(3)=R1*Y(1)*(1.DO-ES)/US
     F(4)=(R3*Y(5)/TCS-R2*Y(4))*(1.DO-ES)/US
     F(5)=-R3*Y(5)/TCS*(1.DO-ES)/US
    RETURN
     END
С
C Subroutine FUNC2
     SUBROUTINE FUNC2(XX,YY,FF)
     IMPLICIT REAL*8(A-H,O-Z)
     COMMON/BLKB/UST, EA
     COMMON/BLKC/R1,R2,R3,CN2
    DIMENSION YY(5), FF(5)
     TCA=YY(1)+YY(2)+YY(3)+YY(4)+YY(5)+CN2
     FF(1)=-R1*YY(1)*(1.DO-EA)/UST
     FF(2)=(R1*YY(1)+2.DO*R2*YY(4))*(1.DO-EA)/UST
     FF(3)=R1*YY(1)*(1.DO-EA)/UST
     FF(4)=(R3*YY(5)/TCA-R2*YY(4))*(1.DO-EA)/UST
     FF(5)=-R3*YY(5)/TCA*(1.DO-EA)/UST
     RETURN
```

```
END
```