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The Advancement of Downdraft Gasification

A thesis presented in partial fulfilment of the requirements for the degree of
Doctor of Philosophy at Massey University

Massey University
New Zealand

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1998

Abstract

Integrated gasification combined cycle systems (IGCC) are currently the most efficient (up to 40 % HHV) thermal method for the conversion of woody biomass to electricity. The production cost of electricity from IGCC is high, U.S. 6.6-8.2 ¢/kW.h (Graig and Mann, 1997). The capital costs alone are U.S. 3.2-4.5 ¢/kW.h. In New Zealand the wholesale price of electricity is typically less than U.S. 2.5 ¢/kW.h.

The bottom line with current IGCC systems is that the capital cost must be greatly reduced if the technology is to be adopted for mainstream use. Methods to reduce the costs of all IGCC components should be investigated.

The use of High Temperature & Pressure (HTP) downdraft technology as an alternative to the current fluidised bed technology could reduce the capital cost of the gasifier component of IGCC by 90%.

The characteristics of the HTP downdraft are different from those of the traditional downdraft. A significant feature is that much larger throughputs are possible at gas turbine combustion pressures. The investigation of HTP downdraft technology is the focus of this thesis.

Acknowledgments

I wish to particularly acknowledge Dr Geoffrey Barnes. As a supervisor, Dr Barnes is enthusiastic and supportive.

I also thank the following:

Dinesh Chand, Electricity Corporation of New Zealand.

Roger Fairclough, East Harbour Management Systems.

Steve Goldthorpe, Woodward-Clyde.

Dr. Josephine Serrallach, Commercial Manager, Massey University.

Dr Francis Thio, Albany Campus, Massey University.

Dr. Tony Clemens, CRL

Colin Wheeler, Two Wheels Engineering

Prof. Murray Hill, former DRC member, Massey University.

Research Services Massey University.

Stevenson's Structural Engineering, Tokomaru.

The former Department of Agricultural Engineering.

Institute of Fundamental Sciences, Massey University.

Table of Contents

ABSTRACT	II
ACKNOWLEDGMENTS	III
1.1 GENERAL INTRODUCTION TO THE ENERGY SECTOR.....	3
1.1.1 <i>Fossil carbon reserves</i>	4
1.1.2 <i>Nuclear power</i>	5
1.2 BIOMASS AS A RENEWABLE CARBON RESOURCE	6
1.2.1 <i>Justification for the use of renewable biomass carbon</i>	6
1.2.2 <i>Current economic uses of bio-energy in New Zealand</i>	7
1.3 AIR GASIFICATION OF WOODY BIOMASS.	8
1.3.1 <i>Comparison of gasification technologies</i>	8
1.3.2 <i>IGCC configurations</i>	10
1.3.3 <i>The importance of developing high pressure gasification technologies</i>	14
1.3.4 <i>Previous work with pressurised downdraft gasification</i>	14
1.3.5 <i>Pressurised fluidised bed gasification for power production, research activities</i>	15
1.3.6 <i>Integrated gasification advanced cycle</i>	17
1.4 PRESSURISED FLUIDISED BED COMBUSTION SYSTEMS	18
1.5 CHEMICAL PROCESSES IN A DOWNDRAFT GASIFIER	20
1.6 MECHANISM OF OPERATION	22
1.7 CHARACTERISING A DOWNDRAFT GASIFIER	23
1.8 HOT GAS CLEANING OF BIOMASS SYNTHESIS GAS.....	25
1.9 REDUCED TAR CONTENT AT HIGHER PRESSURES	27
2.0 INITIAL MODEL USED FOR THE THEORETICAL DEVELOPMENT. 29	
2.1 INTRODUCTION.....	29
2.2 MATHEMATICAL DESCRIPTION OF THE MODEL.....	30
2.3 CONCLUSION	39
3.0 ATMOSPHERIC PRESSURE EXPLORATORY EXPERIMENTS	40
3.1 INTRODUCTION.....	40
3.2 METHOD	40
3.3 RESULTS AND DISCUSSION	44
3.3.1 <i>Pre-steady state results</i>	44
3.3.2 <i>Variation of the frictional function</i>	47
3.3.3 <i>Changes in the bed particle distribution</i>	49
3.4 CONCLUSION	50
4.0 OBTAINING GOOD GASIFIER OPERATION.....	51
4.1 INTRODUCTION.....	51
4.2 METHOD	51
4.2.1 <i>Widening of the gasifier</i>	51
4.2.2 <i>Filtering noise</i>	52

4.2.3 <i>Compaction of the gasifier bed</i>	53
4.3 RESULTS AND DISCUSSION.....	54
4.3.1 <i>Widening of the gasifier</i>	54
4.3.2 <i>Filtering transducer noise</i>	54
4.3.3 <i>Compaction of the gasifier bed</i>	57
4.3.4 <i>Pressure drop mathematical development</i>	59
4.3.5 <i>Increasing the chemical energy content of the fuel-gas</i>	63
4.4 CONCLUSION	66
5.0 GASIFIER SCALE-UP	68
5.1 INTRODUCTION	68
5.2 METHOD	69
5.3 RESULTS AND DISCUSSION.....	69
5.3.1 <i>Pressure effects</i>	69
5.3.2 <i>The correctly operating downdraft gasifier</i>	70
5.3.3 <i>Gasifier reactions</i>	71
5.3.4 <i>The effect of pressure on tar cracking</i>	73
5.3.6 <i>HTP scale-up of the downdraft</i>	74
5.3.7 <i>Dimensional scale-up of the downdraft gasifier</i>	77
5.4 CONCLUSION	81
6.0 FUEL FLEXIBILITY	82
6.1 INTRODUCTION.....	82
6.2 METHOD	84
6.2.1 <i>High moisture content fuels</i>	84
6.2.2 <i>Bark Mulch</i>	86
6.3 RESULTS AND DISCUSSION.....	88
6.3.1 <i>Effect of moisture on the fuel-gas</i>	88
6.3.2 <i>Dry fuels</i>	92
6.3.3 <i>Wet fuels</i>	93
6.3.4 <i>Boosting with natural gas</i>	94
6.3.5 <i>Boosting with coal</i>	95
6.3.6 <i>Black-liquor</i>	95
6.3.7 <i>Bark mulch</i>	97
6.3.8 <i>Bark</i>	98
6.3.9 <i>Coal</i>	99
6.3.10 <i>Start up problems with wet fuels</i>	100
6.3.11 <i>Prevention of start up problems</i>	103
6.4 CONCLUSION	104
7.0 OVERALL CONCLUSION.....	105
8.0 REFERENCES	108
9.0 APPENDIX: GAS CHROMATOGRAPHY DATA.....	114

1.1 General introduction to the energy sector

In today's modern societies energy resources are necessary components for existence. Most of this requirement (85%) is currently supplied by the combustion of fossil carbon reserves (Godley, 1998). The remaining energy needs are supplied predominantly by nuclear and hydroelectric facilities.

Table 1.1 World Energy Requirements. Data from IEO98 (IEO, 1998) and 1997 BP Statistical Review of World Energy (The British Petroleum Company, 1997).

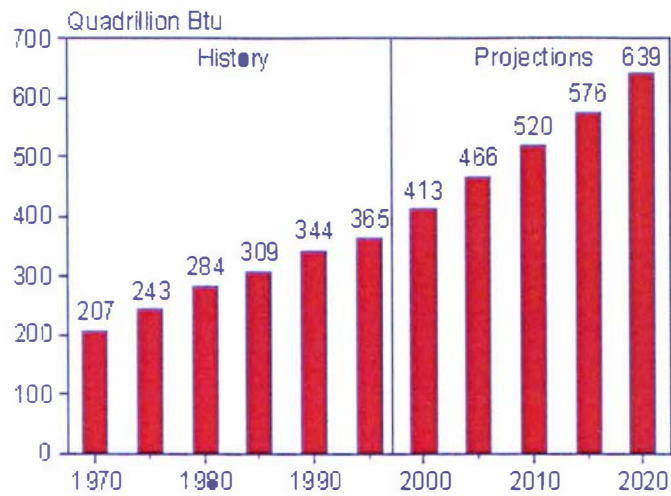
Energy Source	1970	1995	2010	2020	Annual Percent Change		1997 R/P years
					1970-1995	1995-2020	
Oil	97.8	142.5	195.5	237.3	1.5	2.1	40.9
Natural Gas	36.1	78.1	133.3	174.2	3.1	3.3	219
Coal	59.7	91.6	123.6	156.4	1.7	2.2	64.1
Nuclear	0.9	23.3	24.9	21.3	13.9	-0.4	372*
Renewables	12.2	30.1	42.4	50.2	3.7	2.1	Renewable
Total	206.7	365.6	519.6	639.4	2.3	2.3	

*OECD 1993, all land based known and speculative resources

Strong economic growth is expected to substantially boost energy demand over the next two decades. IEO98 projects that total annual world energy consumption could be 75 percent higher in 2020 (Figure 1.1) than it was in 1995 (IEO, 1998).

The majority of this increase in demand (90%) will be supplied using fossil fuels. Most renewable energy development will involve expanding or refurbishing hydroelectric power generation (IEO, 1998). No credible energy forecast predicts that the world will turn away from fossil fuels in the foreseeable future (Godley, 1998).

Figure 1.1



1.1.1 Fossil carbon reserves

There is no current concern that the fossil carbon reserves will be depleted in the near future (50 years). In the early 1990’s concern about theories such as “global warming” caused by the combustion of fossil carbon became an issue. In response to this many countries made a commitment to reduce the consumption of the fossil carbon reserves (Montreal Agreement, United States Global Research Program - National Research Council, 1995).

Significant factual data in support of the “global warming” theory has not been forthcoming. Global temperature has changed little in the last one hundred years. Aircraft laser data indicate that the glacial ice sheets have thickened by approximately 2 meters since observations began in 1980 (Our Changing Planet, 1995). Unless significant scientific data in support of “global warming” or some adverse environmental change is observed, the motivation to reduce fossil fuel consumption may be lost.

Currently, no country (except for countries experiencing decreasing economic activity) is reducing fossil fuel consumption (The British Petroleum Company, 1997). In New Zealand fossil fuel consumption in 1997, had increased 19.4 % since 1990 (The British Petroleum Company, 1997).

The December 1997 Kyoto meeting was an attempt to obtain international agreement over green house gas production. It is doubtful that the outcome of this meeting will provide a mechanism by which fossil carbon consumption will be reduced.

1.1.2 Nuclear power

In 1992 uranium prices had been falling for 14 years. During 1992 the annual spot market price, as indicated by the Nuexco Exchange Value (NEV), reached an all time low of US \$20.67/kg U (Uranium 1993). This equates to a cost of \$2.17/tonne oil equivalent (calculated from the world uranium consumption in Uranium 1997, and the oil equivalent in B.P. Statistical Review of World Energy, 1997). As a result, there has been a substantial reduction of industry employment levels, exploration and new project development.

Growth in nuclear energy consumption has been very strong, 13.9% p.a. since 1975 (Table 1.1). The strong growth is influenced by the extremely cheap cost of the uranium fuel and the zero carbon dioxide emissions, though this is offset by costs of disposal of the nuclear waste and the decommissioning of the nuclear power plants. IEO project that nuclear energy will eventually decline (Table 1.1).

In New Zealand nuclear power is very unpopular because of the fear of nuclear pollution.

1.2 Biomass as a renewable carbon resource

1.2.1 Justification for the use of renewable biomass carbon

Fossil carbon resources have made an important contribution in developing today's high technology world. They have provided an energy source for our machinery and a raw material for many of our organic chemicals requirements. Unfortunately they are a non-renewable resource and therefore will be exhausted eventually. Exhaustion will not occur within our lifetimes, but it is appropriate to develop alternative technologies well before our fossil reserves are exhausted.

Biomass is one of the many alternative energy technologies, which are currently being researched. Furthermore, biomass is a practical renewable carbon resource for our organic chemical materials requirement. Hence the development of renewable biomass technology is an important option for maintaining high technology in future generations.

In some applications, such as in Pulp mills, the use of bio-energy technology is already economically competitive. Therefore it can be argued that further development of this technology could have significant economic benefits.

1.2.2 Current economic uses of bio-energy in New Zealand

1. **The forestry industry.** Waste wood residues are burnt in New Zealand's pulp mills and saw mills. As well as supplying energy, the burning of the waste residues provides a simple method of disposing of the waste and recovering process chemicals (e.g. sodium hydroxide, sodium sulphide, Hotson and Sligh, 1996). The co-generation systems used in the pulping industries are very efficient (up to 90 % NCV, Norris 1996) in that much of the waste heat of electricity production is utilised. *It must be realised that the costs of the waste residues are in effect subsidised by the cost of extracting the valuable wood for pulping and timber production.*

2. **Wood for domestic heating.** It is more economical to directly extract the heat of combustion (high possible efficiency) in the home, rather than convert to electricity (low efficiency) and transmit the power through a grid.

3. **Biogas Technologies.** Biogas is produced from anaerobic decomposition of waste. It is a combustible gas containing mostly methane and carbon dioxide. These technologies are used in New Zealand's meat, wool scour, dairy, vegetable, rendering, pulp and paper, farm manure and sewage sectors (Campbell and Cohen, 1996).

4. **Landfill gas.** Landfills produce a biogas from refuse containing about 55 % methane and 45 % carbon dioxide. Landfills in New Zealand are estimated to generate about 240 million m³ of landfill gas per year. Methane has a global warming potential 21 times that of carbon dioxide. Hence the landfill gas has a greenhouse gas equivalent of 5.65 million tonnes of carbon dioxide (Stewart, 1996). This compares with the total of 3.26 million tonnes of carbon dioxide

emitted from all thermal electricity generation sources in New Zealand (Van der Voorn, 1996).

1.3 Air gasification of woody biomass.

Wood air gasification involves the thermochemical reaction of wood with a limited supply of air to form a combustible mixture of gases (Reed T.B, 1981).

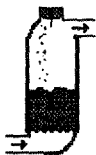


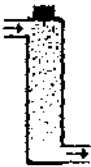


The fuel-gas produced is also known as generator gas and has a very low energy content, typically only about 100-150 kJ.mol⁻¹ HHV.

1.3.1 Comparison of gasification technologies

There are many types of gasifiers that have been designed; some of the more common types are shown in Table 1.3.1 below.

Table 1.3.1 Properties of gasifiers

gasifier design	fuel size (cm)	offgas temp (°C)	tar content (%)	output/diameter
updraft 	1 - 10	400	10 - 30	low
downdraft 	1 - 10	800	0.1 - 1	low
fluidised bed 	0 - 1	800	0.15* - 5	high
entrained flow 	0 - 0.1	1000	0.1 - 1	very high

Reed T.B., Basic chemistry of Gasification, 1980

* Graig and Mann, 1997

Of the gasifiers designed, the entrained flow system has the highest gas output with only low tar content. However it is severely disadvantaged by the requirement for such small fuel size. The fluidised bed gasifiers utilise easily producible fuel size with good gas output; hence these systems have become

very popular (Toft and Bridgewater, 1996). However entrained particulate content and capital costs are substantial.

Downdraft gasifiers can be run on an easily producible fuel size, but they have close specification on the fuel feed (Toft and Bridgewater, 1996). They have a simple robust construction, with high carbon conversion, low tar production and low ash carry over (Toft and Bridgewater, 1996). Historically low output downdraft gasifiers have been used during times of fuel shortage as mobile installations for motor vehicles because of their simplicity and better gas quality. For medium to large gas outputs this system has been abandoned due to the low output of the downdraft gasifier and the dimensional difficulties encountered when attempting to scale up (Kurkela *et al.* 1993).

This PhD research program was focused on the advancement of downdraft gasifier technology, with an objective of overcoming the traditional problems of low throughput and poor fuel flexibility.

1.3.2 IGCC configurations

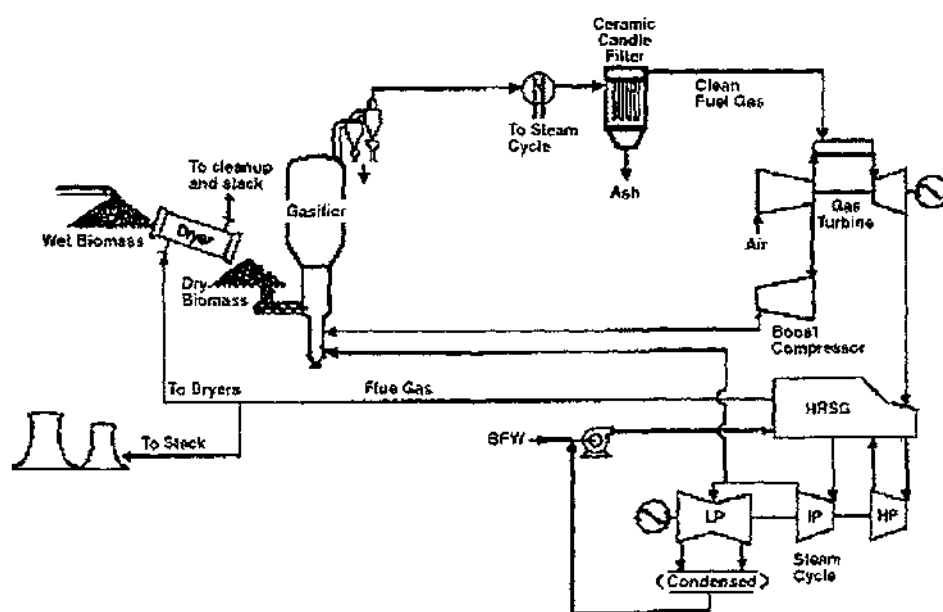
IGCC integrates gasification, gas turbine and steam turbine technology. There are a variety of different IGCC configurations. Three of the common systems are as follows:

1. High pressure IGCC

In high pressure IGCC, the gasifier is operated at a pressure above that of the gas turbine combustor. This process uses both the chemical and sensible energy produced in the gasification process.

A typical high pressure IGCC system is shown in Figure 1.2. In this process air is compressed through the adiabatic compressor of the gas turbine. During compression the air temperature increases to approximately 800 K. Part of the warm (800 K), high pressure, air then passes into the gasifier. After gasification of the air and biomass, the product fuel-gas is then cooled to 800 K before passing through ceramic filters to remove entrained char and ash. The clean fuel-gas then combusts with more air in the gas turbine combustor. Heat is recovered from the gas turbine exhaust and is used to generate steam. The steam is used to dry the wet biomass fuel and to generate additional power.

Figure 1.2 High pressure IGCC (Graig and Mann, 1997)



2. Low pressure IGCC

In low pressure IGCC, the fuel-gas is produced at slightly above atmospheric pressure. A typical example of a low pressure IGCC system is that offered by TPS (Figure 1.3). After gasification, the fuel-gas passes through a tar-cracking reactor. The gas is then cooled to about 561 K (288 C) before filtering using bag filters. The heat generated from the cooling process is used to pre-heat the air used in gasification.

After filtration the fuel-gas is further cooled (to 370 K) and cleaned using a wet scrubber. The cold clean gas is then compressed and fired into the gas turbine.

Figure 1.3 Low pressure IGCC (Graig and Mann, 1997)

3. Low pressure indirect IGCC

Batelle Columbus Laboratory have developed a fluidised bed gasifier that uses circulating hot sand to supply the heat necessary for the endothermic gasification reactions. The sand is circulated between a fluidised bed combustor (which provides the necessary heat) and the gasifier (Figure 1.4). Air is not introduced into the gasifier. The chief advantage of this system is that a fuel-gas with a higher heating value of 16.4 MJ.Nm^{-3} may be produced.

Figure 1.4 Low pressure indirect IGCC (Craig and Mann, 1997)

1.3.3 The importance of developing high pressure gasification technologies

High pressure gasification is an essential component in high pressure IGCC, which has the highest efficiency of all of the IGCC technologies. Pressurised gasification also has thermodynamic advantages over atmospheric gasification in synthetic chemical production and grid piping. In these situations oxygen, steam or hot sand is used as a gasification agent, producing a gas with a higher calorific value. Pressurising these gasification processes makes use of the large molar expansion that occurs in gasification, saving on downstream compression costs (Reed, 1981, p191). It is speculated that the technology can also be used in combination with fuel cells to achieve biomass to electricity efficiencies in excess of 60 % (Makinen *et. al.*, 1994).

While pressurised gasification technology has many technical advantages, it currently is uneconomical for use with biomass. The costs must be greatly reduced if the technology is going to impact upon the current bio-energy market.

1.3.4 Previous work with pressurised downdraft gasification

Toft and Bridgewater (1996) describe the downdraft gasifier as having a “low specific capacity” with “very limited scale-up potential”. Therefore, little research activity in the 90’s has been focused on downdraft gasification (World gasification activities Toft and Bridgewater, 1996), and none on pressurised downdraft gasification for use in combined cycle systems.

Some research was done by SERI in the early eighties with oxygen feed pressurised downdraft gasification. However, their reasons for pressurisation were related to the downstream production of synthetic fuels (Technical and Economic Assessment of Liquid Fuel From Biomass, Dec. 1982). No attempt was made to increase the gasifier output at higher gasifier pressures (Reed and Markson, March 1982). The system they designed was limited by the fuel feed rate.

1.3.5 Pressurised fluidised bed gasification for power production, research activities.

Current research into pressurised gasification of biomass has been limited to fluidised bed (FB) and circulating fluidised bed (CFB) technology. Table 1.2 represents the pressurised gasification activities for power generation. Notice in Table 1.2, that very few are actually operational.

Table 1.2 Research activities into pressurised gasification of biomass

Organisation	Gasifier	Technology	Generator	Status	Output (MW)
Bioflow ¹	Ahlstrom	Pressure CFB	Gas turbine	Operational	6-electrical
PICHTR ¹	IGT	Pressure Air/ O ₂ FB	Not specified	Operational	2-3, electrical
Vattenfall ¹	Tampella	Pressure FB	Gas turbine	Abandoned	60, electrical
Cratech ²	BIG/GT	Pressure FB	Gas turbine	Design	1, electrical
VTT ³	-----	Pressure FB	Not specified	Pilot plant	0.4, thermal
Enviropower ⁴	IGT	Pressure FB	Gas turbine	Design	15, thermal
BIOCYCLE ⁵	-----	Pressure FB	Gas turbine	Design	18, thermal
Delft Uni. ⁶	-----	Pressure FB	Not specified	Pilot plant	1.5, thermal
Lund Uni. ⁷	-----	Pressure CFB	Not specified	Pilot plant	0.1, thermal

FB - Fluidised bed

CFB - Circulating fluidised bed

1 - Toft and Bridgewater, 1996

2 - Craig & Purvis, 1995

3 - Kurkela *et al.*, 1993

4 - Salo *et al.*, 1996

5 - Luxhoi & Madsen, 1996

6 - Andries & Hein, 1996

7 - Wallin *et al.*, 1996

1.3.6 Integrated gasification advanced cycle

Integrated gasification advanced cycle (IGAC) systems utilise a single, advanced, moisturised gas turbine cycle or other turbine system improvements resulting from the Advanced Turbines Systems program (DOE³, 1998). In a moisturised gas turbine cycle, the gas turbine replaces the need for an additional steam turbine by passing the steam from the recovery boiler (recuperator) into the gas turbine. In other words the moisturised gas turbine cycle is effectively both a gas turbine and a steam turbine in one. By eliminating the steam turbine from the IGCC cycle, capital cost is reduced.

Historically coal and biomass IGCC systems have used retrofitted methane gas turbines. By designing gas turbines specifically for coal and biomass fuels, better efficiencies are expected. Gas turbine improvements being developed for coal and biomass applications include (DOE¹, 1998):

- Rich-quench-lean combustors that reduce NO_x emissions
- Hot gas scrolling between the combustor and the turbine expander
- Combustors that accept gas with a range of compositions and heating values
- Techniques to extract air from the compressor discharge with minimum pressure loss

The U.S. DOE strategic objective is to achieve 50% net efficiency from IGAC by the year 2010 (DOE³, 1998).

1.4 Pressurised fluidised bed combustion systems

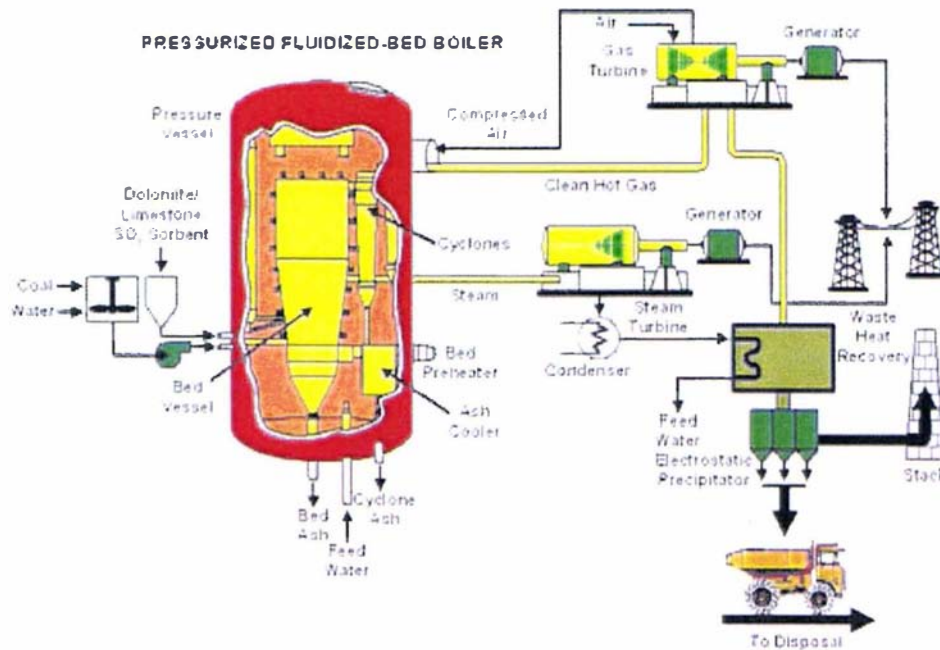
Pressurised fluidised bed combustion (PFBC) is an efficient combustion alternative to IGCC. PFBC is not the focus of this thesis, however since PFBC is a competition technology, a brief description is given.

High efficiency, high pressure, combustion systems are being developed. These systems use a pressurised fluidised bed combustor (PFBC) to produce steam to power a steam turbine, which is the primary source of power. An example of this is shown in Figure 1.5. The pressure is supplied from a gas turbine operating at a temperature of 870 °C. At these low temperatures the available power from the gas turbine is derated.

Current status is that this technology is still being held up by hot gas cleaning difficulties. The basic problem is that the filters become embrittled at higher temperatures. This causes the performance to drop off and the filters break apart. To quote from the DOE's PFBC website (DOE⁴, 1998), **“Further development of ceramic barrier filters is urgently needed”**. It has been on the urgent list for a long time now. A large portion of the 1999 funding for PFBC has been allocated to advancing hot gas cleaning (Godley, 1998).

If the cleaning difficulties can be overcome, it is speculated that these first generation systems will have an efficiency of 42%. However, after many years of effort, and huge investment, it is now less certain that these embrittlement problems can be easily solved.

Figure 1.5 PFBC



The second generation systems use a process called topped PFBC (DOE⁴). In this system a carbonizer is introduced. A carbonizer is a gasifier that produces fuel-gas and char. The char is used in the combustor. The fuel-gas is combusted in the gas turbine to increase the temperature to the rated gas turbine specifications. Power obtained from the gas turbine is increased and power from the steam turbine is decreased.

The second generation system has the appearance of quasi IGCC technology. If this technology were applied to biomass, it would be called IGCC because 85% of biomass is volatiles; hence only a minority of the energy would end up in the PFBC. This side-step may allow PFBC developers to get around the problem of high temperature filter embrittlement by filtering at a lower temperature, then increasing the temperature again by firing with fuel-gas. The combining of PFBC, carbonizer, steam turbine, and gas turbine technology, does add to the complication of the process.

The DOE have set a second generation strategic objective of 45% efficiency by 2010 (DOE⁵, 1998). This is a less optimistic objective than for the IGAC

systems (50% by 2010). Furthermore the IGAC systems are considerably simpler, combining only a gasifier and a gas turbine.

On a more philosophical level, PFBC is a fascinating research development. PFBC is in theory a good mechanism by which combustion technology may be advanced. It is hoped that the urgent problem of filter embrittlement is resolved.

1.5 Chemical processes in a downdraft gasifier

In the downdraft gasifier there are three basic processes occurring; combustion, pyrolysis and gasification (Figure 1.6).

(1) Combustion involves the exothermic (heat producing) reaction of oxygen (from the air) with the biomass to form oxygenated products such as H_2O and CO_2 .

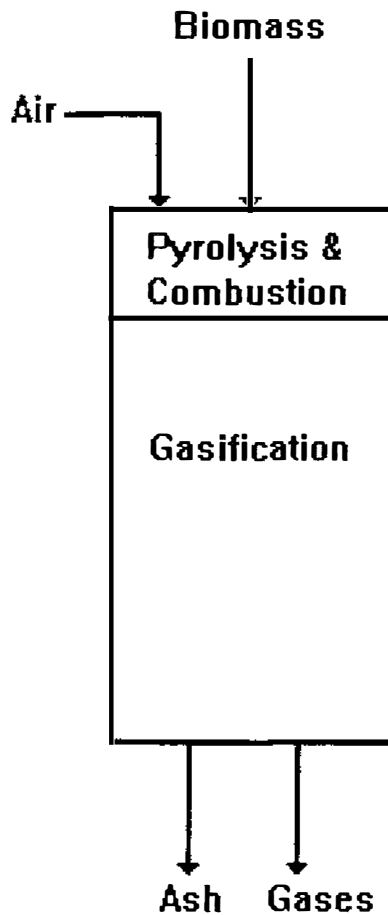
(2) Flaming pyrolysis is the thermally induced breakdown of biomass to form volatile combustible gases (75-85 % by mass). The combustible pyrolysis gases are then thermally cracked, combusted and gasified to form fuel-gas.

(3) Gasification involves the endothermic reduction of H_2O (equation 1.5.1) and CO_2 (equation 1.5.2) by a carbon source to form a combustible mixture of gases.

Water-gas reaction (equation 1.5.1) $\text{H}_2\text{O} + \text{C} \rightarrow \text{CO} + \text{H}_2$ 136.0 kJ.mol⁻¹

Boudouard reaction (equation 1.5.2) $\text{CO}_2 + \text{C} \rightarrow 2\text{CO}$ 170.7 kJ.mol⁻¹

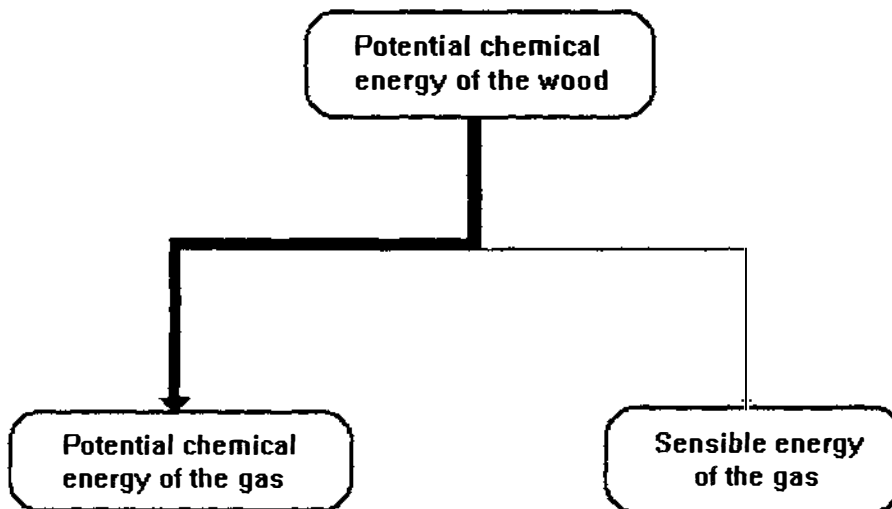
Figure 1.6



Gasification reaction rates are about 240 times slower than combustion reactions at 900 °C (Graboski, 1981). At higher temperatures (above 1400 °C) gasification reaction rates become so rapid that they moderate the temperature preventing further temperature increase from combustion. A dramatic example of this can be seen for the downdraft gasifier. If pure oxygen is used the temperature does not rise more than one hundred degrees above that of using just air, except in the immediate vicinity of the nozzles (Desrosiers, 1981).

The thermo-chemical energy balance of a gasifier can be represented by Figure 1.7, assuming that the heat losses from the gasifier bed are minimal and there is negligible char production.

Figure 1.7



In the gasification process the potential chemical energy of the biomass fuel is transformed into potential chemical energy of the fuel gas and sensible energy. The sensible energy represents both temperature volume changes and molar volume changes. For high pressure IGCC systems both the chemical energy and the sensible energy of the gasification are utilised for power production.

1.6 Mechanism of operation

In the downdraft gasifier, fuel and air are reacted as the air passes downwards through a fixed bed of carbonised fuel particles:

- Fuel feed is by gravity bulk solids flow.
- Air is fed through the bed from nozzles in the throat.

In order to get greater throughput out of a given reactor; it is necessary to pass more air and more fuel into the reactor. What limits this?

1. Fuel feed rate by gravity has to be adequate. This is a function of the design of the hopper outlet and the nature of the bulk materials. Regarding fuel flow properties:
 - Free flowing material normally can flow into the reaction zone at a greater rate than the required throughput – therefore no limitation.
 - Some materials e.g. shredded stringy bark, may not flow freely enough and this will be a limit to operation.
 - Some materials may sticky during the drying and pyrolysis stages and this could interfere with flow.
2. Ability to get the air to flow through the bed at the required rate:
 - There is a practical limit to the flow velocity through the packed bed.
 - High velocities would lead to high pressure drops, and would likely compact soft char particle in the bed, causing a blockage
 - Operating at higher pressure would allow higher molar flow rates through the bed with acceptable pressure drop. This is demonstrated by the mathematical model in chapter 2.
3. The kinetics of the reactions. The kinetic reaction rate is influenced by:
 - The gasification temperature.
 - The partial pressure of the reactive gases.
 - The delivery of the reactive gases to char active sites.
 - The char surface area available.

1.7 Characterising a downdraft gasifier

Gasification relies on reacting air and fuel together in proportions that are sub-stoichiometric for complete combustion. Equilibrium calculations and experimental investigations suggest that the optimum product gas heating value can be obtained when the equivalence ratio, Φ is between 0.25 and 0.4.

$$\Phi = \frac{A/F_{\text{Actual}}}{A/F_{\text{stoichiometric}}}$$

For a given bed geometry (i.e. bed diameter, bed length) it should be possible to determine the gas quality and operating temperature as a function of the air flow rate (or gas production rate). This will depend on the flow of the fuel not being restricted by poor flow properties.

Supply of fuel to the reaction zone is driven by the gravity feed of biomass fuel into the bed of the gasifier. This in turn is restricted to the rate of removal of material from the bed by conversion of the solid material to gas and ash, with removal of ash through the grate.

Given the geometry, and an unrestricted fuel flow, it should be possible to characterise the gasifier behaviour in terms of the gas heating value and temperature as functions of airflow under steady state conditions. One would expect to obtain an optimum airflow rate for a particular fuel and gasifier geometry.

With this basic characterisation, the optimum operating flow rate can be defined and from this the thermal capacity of the gasifier in terms of grate area determined. This could be called the heat-loading rate, defined as:

$$HR = \frac{\text{Heat released from fuel (MW}_t\text{)}}{\text{Throat area (m}^2\text{)}}$$

The geometry is known to influence performance of the gasifier, with many reports indicating the need to adjust bed diameter, bed length, and air injector

arrangements. The optimum geometry is commonly developed on a trial and error basis, after using crude measures of gas heating value.

The effect of pressure can also be studied in this way. It should be possible for a given geometry to find the optimum molar flow rate of air at each different pressure and thus establish the throughput and the heat rate as functions of pressure.

Similarly the effect of changes in the fuel composition can be studied. The major influence (for woody biomass) in this study is clearly the influence of moisture, because of the large heat impact of evaporation.

1.8 Hot gas cleaning of biomass synthesis gas

During the process of gasification entrained ash and char particulates are produced, which if not removed cause problems of erosion, corrosion and deposition of/in the turbine blades in a gas turbine.

The cleaning of biomass fuel-gas can be divided into two general categories; hot gas cleaning and cold gas cleaning. In cold gas cleaning the gas is cooled via heat exchangers down to an ambient temperature which causes much of the water and tar to condense. The remaining tar droplets, char, and ash are removed via filtration. In hot gas cleaning, the char and ash are removed via filtration while the gas is hot. No condensation of the tar and water occurs. Hot gas cleaning (HGC) is used in high pressure IGCC.

The advantages of hot gas cleaning over cold gas cleaning are summarised below:

- (1) Higher achievable efficiencies are possible since the sensible heat of gasification can be best utilised.
- (2) Less heat is removed using heat exchangers.
- (3) No condensate is produced. Since condensate contains many toxic chemicals, hot gas cleaning is better for the environment.

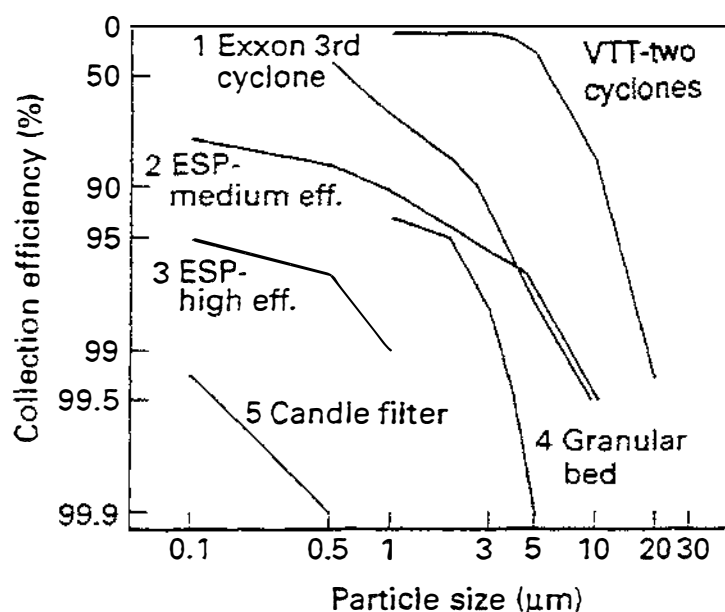
Most components of tar are not a problem in HGC, however the decomposition of these pyrolysis (tar) products leads to a formation of high-molecular-weight components and soot which may cause blinding of the ceramic filters (Kurkela et al, 1993). These problems may be avoided by the use of a tar cracking reactor prior to filtration (Mark & Paisley, 1994). However this may significantly add to the initial and on-going costs of the gasifier installation.

There are four basic methods by which particulates are currently removed from the hot gas stream:

- (1) Cyclonic action
- (2) Electrostatic precipitation (ESP)
- (3) Ceramic candle filters
- (4) Granular bed filters

Typical collection efficiencies of the different collection methods are shown in Figure 1.8 (Kurkela et. al., 1993).

Figure 1.8



Of these systems, ceramic candle filter systems are most favoured because they have the highest collection efficiency. Candle filters are cleaned periodically by back-flushing in pulses with N_2 , CO_2/N_2 or synthesis gas. Another method for cleaning the filters is to entrain air in with the dirty fuel-gas which combusts the cake (Pederson *et. al.*, 1997). The material for ceramic candles is usually silicon-carbide based.

It is recommended that the alkali vapour levels be reduced to below estimated turbine tolerance levels of 50 to 100 ppb (Wiant *et. al.* 1993, Mojtadehi *et.al.*1991, Ye *et. al.*, 1996). To reduce the alkali vapour to acceptable levels it is necessary to filter at temperatures less than 873 K (Kurkela *et. al.*, 1993).

1.9 Reduced tar content at higher pressures

The predominant reason why downdraft gasifiers produce synthesis gas of such low tar content is because they are designed so that all pyrolysis gases (including tars) must pass through a hot zone (up to 1400 °C, Desrosiers, 1981)

where the tar undergoes cracking (Garcia-Bacaicoa *et. al.*, 1995) and subsequent combustion/gasification. If the gasification pressure were increased, the rate at which the tars are cracked must increase also (Rosen *et. al.* 1996, NATO 1985) because the reactions are dependent on the partial pressures of the combustion gases (e.g. steam). Hence, in the oil industry, all hydro and steam crackers are operated at elevated pressures.

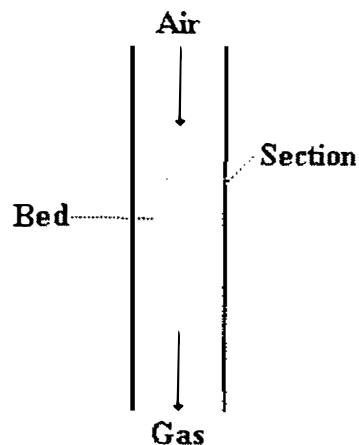
2.0 Initial model used for the theoretical development

2.1 Introduction

A theoretical model was developed to describe the physical flow of gases through a downdraft gasifier bed. A downdraft gasifier can be represented by Figure 2.1. Air enters evenly into the top of the gasifier bed. As the gas stream passes through the bed, combustion increases the temperature to approximately 1600 K. At this point reduction reactions begin to dominate, reducing the temperature to about 1000-1400 K before the gas stream exits the bed. Throughout this entire process molar expansion of the gas stream is occurring.

The volume of the bed can be divided into “n” horizontal sections, where “n” is large enough that only small temperature and molar changes occur over the length of each section.

Figure 2.1



In high pressure IGCC system, air is compressed adiabatically into the gasifier. Adiabatic compression results in a simultaneous increase in both the pressure and the gasifier intake air temperature. So that these effects can be studied independently of one another, an isothermal compressor will be used to study the effect of pressure alone.

2.2 Mathematical description of the model

If each of the sections (Figure 2.1) is considered as having gas flow properties which are analogous to a typical bed of solids, then the friction drag force per unit area of the section can be given by equation 1 (McCabe, 1976).

$$\frac{F_D}{A_{ss}} = \frac{\rho_{ms} u^2 k_2}{g_c} \quad (1)$$

where

F_D = frictional drag force (N) through the bed

A_{ss} = surface area of the char particles in the section (m^2)

ρ_{ms} = mass density of the gas ($g.m^{-3}$)

u = velocity of the gas ($m.s^{-1}$)

k_2 = constant for the system

g_c = Newton's law proportionality factor for the gravitational force constant

Rearranging (1)

$$F_D = \frac{A_{ss} \rho_{ms} u^2 k_2}{g_c} \quad (2)$$

The power loss (P , $J.s^{-1}$) due to the friction against gas flow of the section is given by

$$P = F_D u \quad (3)$$

Combining (2) and (3)

$$P = \frac{A_{ss} \rho_{ms} u^3 k_2}{g_c} \quad (4)$$

The velocity is equal to the volume flow rate in the section (v_s , $m^3.s^{-1}$) divided by the cross-sectional area of the channels in the section (A_{cs} , m^2).

$$u = \frac{v_s}{A_{cs}} \quad (5)$$

Substituting (5) into (4)

$$P = \frac{A_{ss} \rho_{ms} v_s^3 k_2}{A_{cs}^3 g_c} \quad (6)$$

The molar work (w_s , $J.mol^{-1}$) for the frictional losses in the section is given by equation (7).

$$w_s = \frac{P}{f_s} = \frac{A_{ss} \rho_{ms} v_s^3 k_2}{A_{cs}^3 f_s g_c} \quad (7)$$

where

f_s = molar flow rate (mol.s^{-1}) in the section

It is assumed that pressure drop through the bed is small relative to the gasifier bed pressure.

$$\Delta p_b \ll p \quad \therefore p_i \approx p_s \approx p_f \approx p \quad (8)$$

where

Δp_b = pressure drop through the bed (Pa)

p_i = input air pressure (Pa)

p_s = section air pressure (Pa)

p_f = exit gas pressure (Pa)

p = is the pressure of the gasifier bed (Pa)

Taking the ideal gas equation

$$n = \frac{pV}{RT_i} \quad (9)$$

where

V = volume of gas (m^3)

R = $8.31441 \text{ m}^3.\text{Pa}.\text{K}^{-1}.\text{mol}^{-1}$

p = pressure of the gasifier (Pa)

Noting that the input volume flow rate (v_i) is equal to the volume flowing through the section divided by the time taken.

$$v_i = \frac{V}{t} \quad \therefore V = v_i t \quad (10)$$

where

t = time (s)

Substituting (10) into (9) and dividing by time.

$$\frac{n}{t} = \frac{p_i v_i}{RT_i} = f_i \quad (11)$$

where

f_i = molar flow rate of air into the gasifier (mol.s^{-1})

It should be noted that the word “throughput” is a relative term, used to describe the flow of gas (air) into the gasifier, passing through the bed, and finally exiting as generator gas. Relative increases in throughput can be described either in terms of exit flow rates or as input flow rates.

An analogous equation for the molar flow rate in the section is given in equation (12).

$$\frac{n}{t} = \frac{p_s v_s}{RT_s} = f_s \quad (12)$$

where

f_s = molar flow rate of air in the section (mol.s^{-1})

The section flow rate (f_s , mol.s^{-1}) is equal to the input flow rate multiplied by the molar expansion occurring in the bed up to the section (X_s).

$$f_s = f_i X_s \quad (13)$$

Substituting (12) into (13)

$$\frac{p_s v_s}{RT_s} = \frac{p_i v_i X_s}{RT_i}$$

$$\frac{v_s}{T_s} = \frac{v_i X_s}{T_i} \quad \text{since } p_i \approx p_s$$

$$v_s = \frac{v_i T_s X_s}{T_i}$$

where

X_s = molar expansion that has occurred up to the section

v_i = input volume flow rate of air ($\text{m}^3 \cdot \text{s}^{-1}$)

T_i = input temperature of the air (K)

T_s = section temperature of the gas (K)

Rearranging

$$v_i = \frac{f_i RT_i}{p_i} \quad (14)$$

Substituting (14) into (13)

$$v_s = \frac{f_i RT_s X_s}{p_i} \quad (15)$$

Substituting (15) into (7)

$$w_s = \frac{A_{ss} \rho_{ms} f_i^3 R^3 T_s^3 X_s^3 k_2}{A_{cs}^3 g_c f_s p_i^3} \quad (16)$$

Substituting (13) into (16)

$$w_s = \frac{A_{ss} \rho_{ms} f_i^2 R^3 T_s^3 X_s^2 k_2}{A_{cs}^3 g_c p_i^3} \quad (17)$$

The mass density of the gas in the section is equal to the molar density of the gas multiplied by molar mass of the gas.

$$Mr_{gs} = \sum_{\text{For all components}} Mr(\text{component}) \times \text{mol fraction}(\text{component})$$

$$\rho_{ms} = \rho_{ns} Mr_{gs} \quad (18)$$

where

ρ_{ns} = molar density of the gas in the section (mol.m^{-3})

Mr_{gs} = relative molecular mass of the gas in the section (g.mol^{-1})

The ratio of relative molecular masses between gas in the section and air is given as.

$$Y_s = \frac{Mr_{gs}}{Mr_{air}} \quad \therefore Mr_{gs} = Y_s Mr_{air} \quad (19)$$

where

Y_s = ratio of relative molecular masses of the gas in the section and air

Mr_{air} = molecular mass of air ($=28.964 \text{ g.mol}^{-1}$, Weast, 1976-1977)

The ideal gas equation can be expressed as:

$$\frac{p}{R} = \frac{nT}{V} = \rho T$$

At constant pressure, the molar density in the section can be related to the input molar density by equation (20).

$$\rho_{ns} T_s = \rho_{ni} T_i$$

$$\rho_{ns} = \frac{\rho_{ni} T_i}{T_s} \quad (20)$$

$$\rho_{ni} = \frac{n}{V} = \frac{p_i}{RT_i} \quad \text{ideal gas equation} \quad (21)$$

Substituting (21) into (20)

$$\rho_{ns} = \frac{p_i}{RT_s} \quad (22)$$

Substituting (19) and (22) into (18)

$$\rho_{ms} = \frac{p_i Y_s M_{r_{air}}}{RT_s} \quad (23)$$

substituting (23) into (17)

$$w_s = \frac{A_{ss} f_i^2 R^2 T_s^2 X_s^2 Y_s M_{r_{air}} k_2}{A_{cs}^3 g_v p_i^2} \quad (24)$$

The molar work for the bed (w_b) is equal to the summation of the molar work for each section.

$$w_b = \sum_{s=1}^n w_s$$

From equation (23)

$$w_b = \sum_{s=1}^n \frac{A_{ss} f_i^2 R^2 T_s^2 X_s^2 Y_s M_{r_{air}} k_2}{A_{cs}^3 g_c p_i^2} \quad (25)$$

The friction function “C” of the gasifier bed may be defined as.

$$C = \sum_{s=1}^n \frac{A_{ss} R^2 T_s^2 X_s^2 Y_s M_{r_{air}} k_2}{A_{cs}^3 g_c} \quad (26)$$

The numerical value of the friction function can be used as a measure of gasifier throughput capacity. The higher the friction function the lower the throughput capacity.

Substituting (26) into (25)

$$w_b = \frac{C f_i^2}{p_i^2} \quad (27)$$

The work done forcing each mole of gas through the section is supplied by the compressor. For these studies an isothermal compressor will be used. Isothermal compression has the advantage that the effects of gasifier pressure and intake air temperature can be studied separately.

The molar work of isothermal compression in the section (w_s) is given by-

$$w_s = RT_i \ln \left(\frac{p_b}{p_e} \right) \quad (\text{Atkins, 1988}) \quad (28)$$

where

p_b = pressure at the beginning of the section (Pa)

p_e = pressure at the end of the section (Pa)

The total molar work done on the bed by the compressor is equal to the summation of all the work done in each section.

$$w_b = \sum_{s=1}^n RT_i \ln \left(\frac{p_b}{p_e} \right) = RT_i \ln \left(\frac{p_i}{p_f} \right) \quad (29)$$

Combining (26) and (28)

$$w_b = RT_i \ln \left(\frac{p_i}{p_f} \right) = \frac{C f_i^2}{p_i^2} \quad (30)$$

If the friction function “C” is constant for variable molar flow rate and pressure then the simple solution (equation 31) can be obtained.

$$w_b \propto \frac{f_i^2}{p_i^2} = \left(\frac{f_i}{p_i} \right)^2 \quad (31)$$

Equation (31) predicts that the energy requirements (work) per mole of gas, increase with the square of the molar flow rate, and decrease with the square of the pressure. If the pressure were increased 25 fold (i.e. gas turbine pressures) then the flow rate could be increased 25 fold without increasing the energy

requirements (work). Note this analysis has not considered any kinetic changes. It describes only energy loss as a result of physical flow and pressure in the bed.

2.3 Conclusion

A mathematical model of the downdraft gasifier has been developed. It was assumed that the downdraft gasifier bed behaves similarly to a typical bed of solids. From this analysis it is predicted that elevated pressure may be used to greatly scale up the throughput of the downdraft gasifier. This analysis does not include kinetic considerations.

3.0 Atmospheric pressure exploratory experiments

3.1 Introduction

The initial exploratory experiments were designed to rapidly explore the effects of volume flow rate on the molar work of compression at various pressures. This information was to be used to test the mathematical model developed in Chapter 2. The theory had predicted a squared relationship between the input molar flow rate and the molar work. Furthermore it had predicted that as the pressure increased the molar work would decrease.

3.2 Method

A high pressure downdraft gasifier was designed (Figures 3.1.1 and 3.1.2). The device was manufactured initially by myself at Two Wheels Engineering, Palmerston North. Subsequent modifications such as the larger hopper were made by Russell Watson at Agricultural Engineering, Massey University.

The gasifier shell was constructed from CNG tanks, which are designed to cope with pressures up to 12000 kPa. The shell was insulated from the gasifier bed by a layer of high temperature Kao-Wool insulation. The gasifier throat was constructed from fire bricks.

Air was supplied to the bed from 16 x 8 mm nozzles. The grate depth was externally adjustable. The bed diameter at the nozzles, the throat, and the grate, were 160 mm, 120 mm and 200 mm respectively.

Below the gasifier was a crude cyclone, which separated out the char-ash from the gas stream. The char-ash was deposited in a collector constructed from a new 20 l LPG tank. Pressure was maintained in the gasifier by a high temperature exit valve. The whole device was independently pressure tested and certified to 2200 kPa (300 p.s.i, boost) by National Gas (Palmerston North).

Figure 3.1.1 Original gasifier

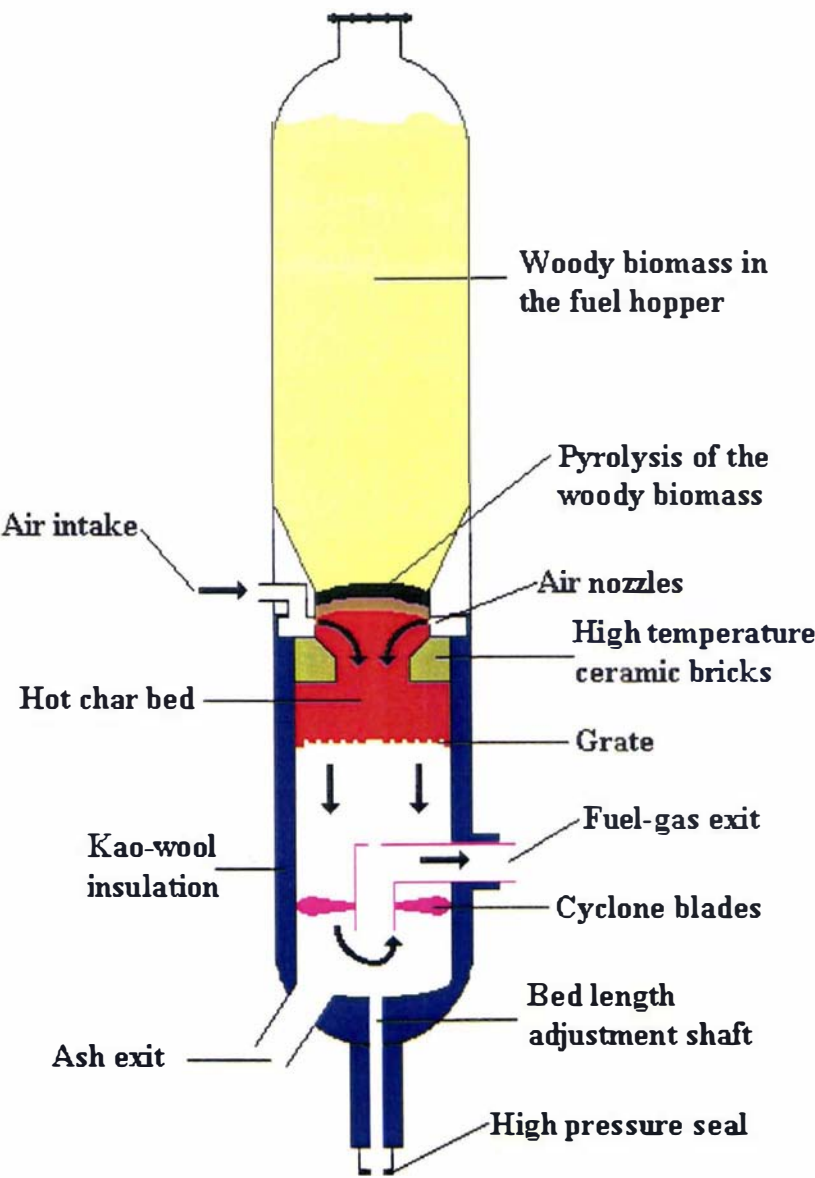


Figure 3.1.2 Gasifier operating at atmospheric pressure in a lecture theatre. PhD candidate Richard Sime is shown on the left. Supervisor Dr. Geoffrey Barnes is depicted on the right.



The air pressure supplied to the gasifier was controlled using a high flow regulator. The fuel-gas was flared off in a crude burner. The gasifier was run at pressures of 130, 200, 360 and 800 kPa, using dry wood-chip (m.c. 4% by weight). The bed length (measured from the air nozzles to the grate) was 10 cm. Readings were made every 0.5 seconds. At each pressure, data were collected over a range of volume flow rates by slowly opening and closing the exit valve.

The experimental parameters monitored using a Campbell 21X data-logger were:

- Intake air temperature; measured using a RS K-type thermocouple, calibrated at 0 °C and 100 °C using ice water and boiling water.
- Intake air flow; measured using an orifice plate flow meter with a Farnell SX01DN, calibrated against axial flow air velocity flow meter
- Gasifier pressure; measured using a Farnell SX150DN pressure transducer, calibrated against a dead weight pressure calibration device.
- Pressure drop through the bed; measured using a Farnell SX01DN pressure transducer, calibrated against a water manometer.
- Fuel-gas exit temperature; measured using a RS N-type mineral insulated thermocouple, calibrated against a McGregor Eurotherm electric furnace thermocouple. The furnace thermocouple had been previously checked using standards with known melting points.

Data recorded when the valve was fully closed was used to zero the bed pressure drop and flow rate transducers. These transducers were prone to drift, hence regular closing of the exit valve allowed better measurement. After the data were collected on the Campbell data-logger, they were transferred to an audio tape cassette. The data were read from the audio tape using an old 286 computer. It was then run through a program to remove unwanted information.

The clean data were stored on a 5.5 inch floppy, and then transferred to a 3.5 inch floppy where they could be read into an excel program for analysis. The whole process was time consuming and the inability to observe the gasifier in real time was a limiting feature for experimental design.

3.3 Results and discussion

3.3.1 Pre-steady state results

The theory developed in chapter 2 predicts a squared relationship between the input molar flow rate of air into the gasifier and the molar work of compression required to overcome the pressure drop through the bed. Figure 3.2 is a graph of the molar work vs the input molar flow rate from data collected at atmospheric pressure. Figure 3.3 is a replot of the data against the input molar flow rate/pressure squared. Note that the relationship is linear as predicted.

Figure 3.2

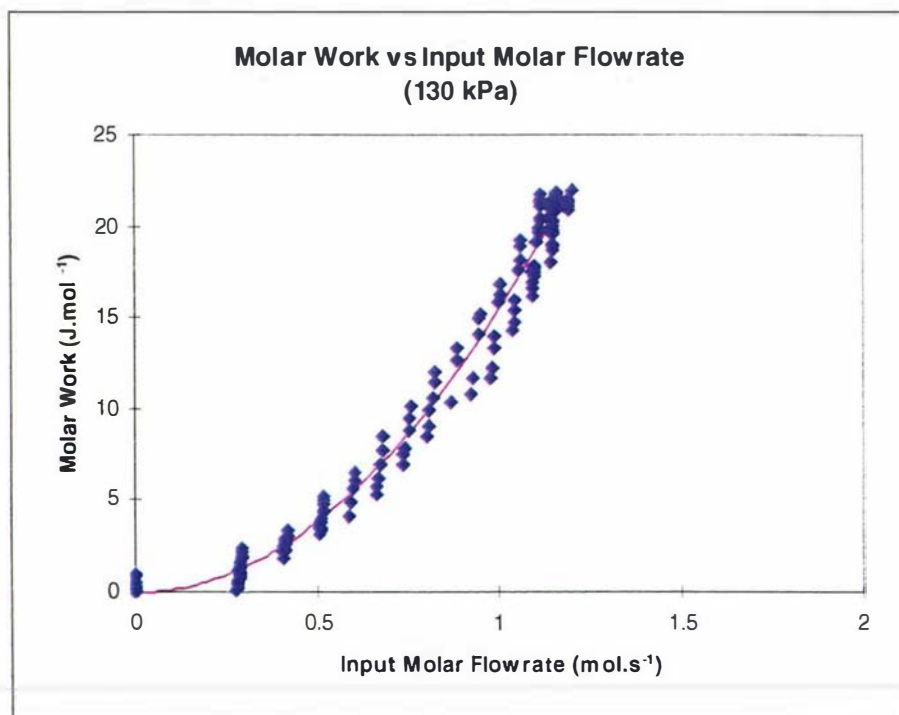
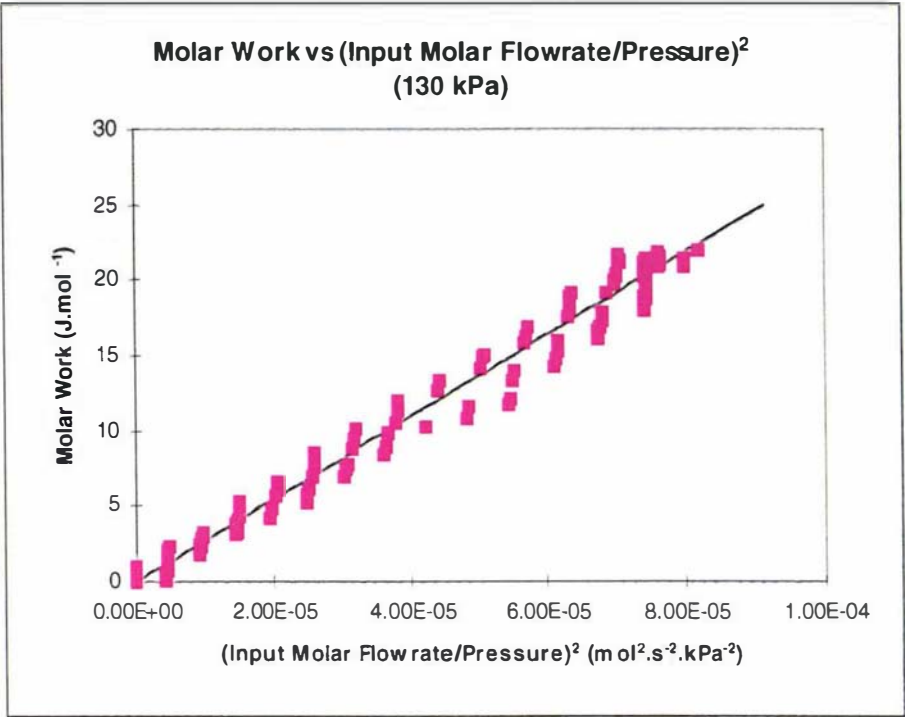
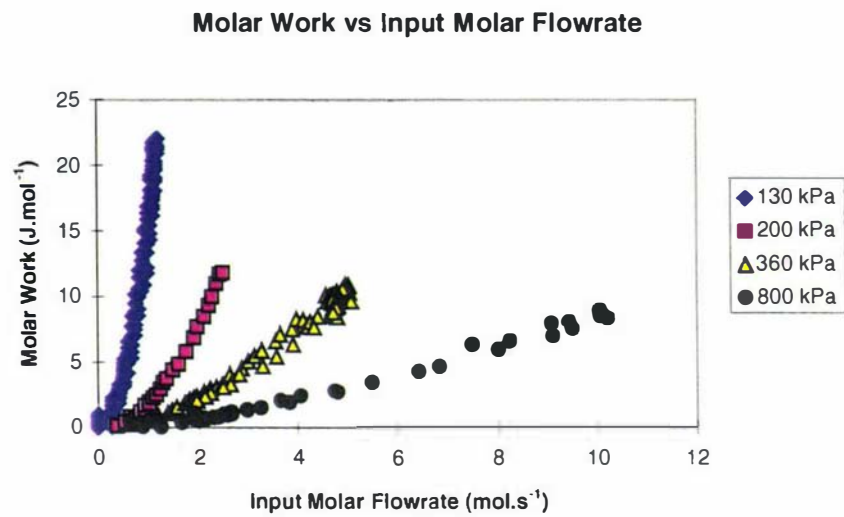


Figure 3.3



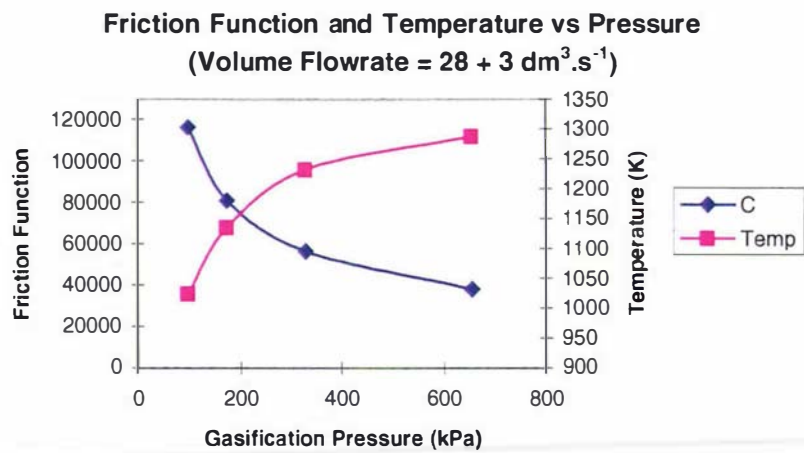
Similar results were obtained at 200, 360, and 800 kPa (Figure 3.4). It can be seen that the molar flow rate of air into the gasifier is increased at higher pressure without increasing the energy loss (molar work of compression). This validates the theoretical principle of the mathematical model developed in chapter 2.

Figure 3.4



The exit temperature increased with increasing pressure (Figure 3.5), indicating that the chemical reaction rate in the bed was not able to keep up with the increased physical flow of gas through the bed

Figure 3.5



3.3.2 Variation of the frictional function

The value of the friction function “C”, decreased with increasing pressure (Figure 3.5). Hence, lower than expected energy losses were observed at higher pressures. A similar observation was made for changes in the volume flow rate, where the frictional function decreases with increasing volume flow rate (Figure 3.6). The temperature record for this data set also showed an increase in fuel-gas exit temperature with increasing volume flow rate (Figure 3.7). Note that the response of both the temperature and the frictional function appear to be slightly delayed from that of the volume flow rate.

Figure 3.6

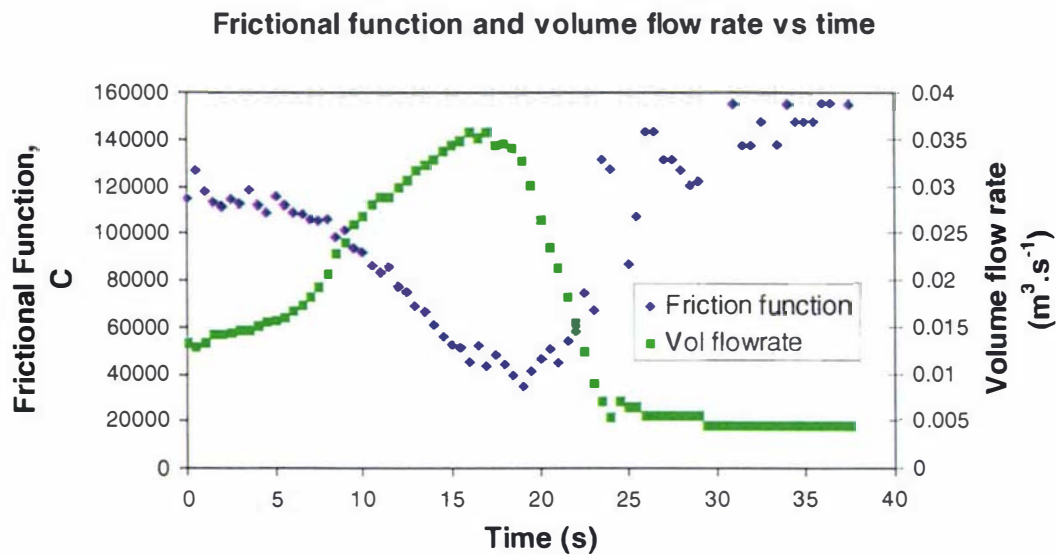
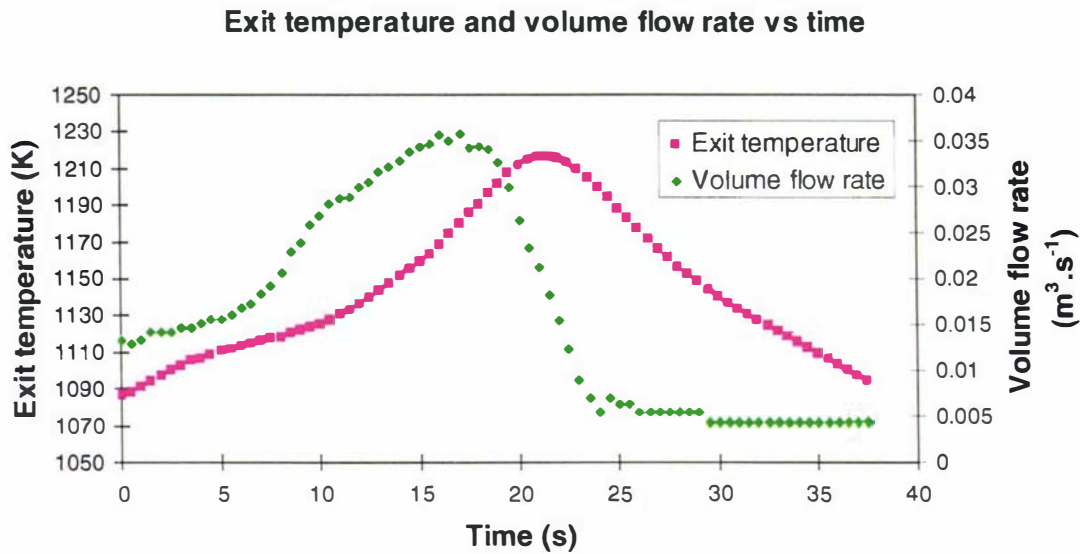


Figure 3.7



A decrease in the friction function was accompanied by an increase in the fuel-gas exit temperature for both increased pressure and increased volume flow rate. Both pressure and volume flow rate contribute to the molar flow rate of air into the gasifier. Therefore it can be said, that increased molar flow rate results in an increase in the exit temperature and a decrease in the frictional function.

From gas chromatography of the composition of the fuel-gas at high and low exit temperatures, it can be shown that the value of T^2X^2Y in equation (26) increases with increasing temperature. Therefore exit temperature and its effect on the fuel-gas composition can not account for the observed decrease in the frictional function

$$C = \sum_{s=1}^n \frac{A_{ss} R^2 T_s^2 X_s^2 Y_s M_{r_{air}} k_2}{A_{cs}^3 g_c} \quad (26)$$

Other parameters, R , M_{air} , k_2 and g_c , are all constants. By the process of elimination the decrease in the friction function with increased molar flow rate can be attributed to changes occurring in the bed particle distribution ($\frac{A_{ss}}{A_{cs}^3}$).

3.3.3 Changes in the bed particle distribution

Changes in the bed particle distribution with temperature can be rationalised from current understanding of char reactivity.

At low temperatures (<1173 K) surface kinetic reactions are slow, all the reactive sites throughout the porous char particle are saturated with reactive gases (Graboski, 1981). Therefore at these low temperatures both small and large char particles are shrinking at the same relative rate. Furthermore the internal structure of the char particle becomes weak, causing the particle to fracture and break up. Note, there is continuing debate over the exact nature of surface kinetics mechanisms (Bhatia, 1998). As this is not important for these discussions such matters will not be entered into.

At high temperatures surface kinetic reactions are fast. All of the reactive gases are exhausted close to the surface of the char particle (Graboski, 1981). The centre of a large particle is unreactive. Under these conditions small char particles shrink relatively faster than the large particles because the small particles have a larger surface/volume ratio. This factor tips the bed distribution in favour of large char particles.

Furthermore the internal structure of char particles under these conditions is not eroded away to the same extent, therefore the particles are less likely to break up. In addition, with high exit temperatures the bed no longer has a dead zone. The dead zone in a downdraft gasifier is prone to blocking with small particles.

These particles can not be chemically removed because insufficient energy remains. To remedy this problem, downdraft gasifiers are often equipped with automated char removal systems, which physically remove the fine char build up from the dead zone.

The combination of all these factors cause the gas stream to flow more freely at high exit temperatures. Thus, as observed, the frictional function decreases at high exit temperatures.

3.4 Conclusion

The data collected in these experiments may be regarded as pre-steady state (non-equilibrium). These measurements should be accompanied with steady state measurements. To achieve this, real time monitoring of the gasifier is essential.

Changes in the bed particle distribution at increased molar flow rate were observed. While this is an advantageous feature in terms of scale up, it does add to the difficulty of modelling the process.

4.0 Obtaining good gasifier operation

4.1 Introduction

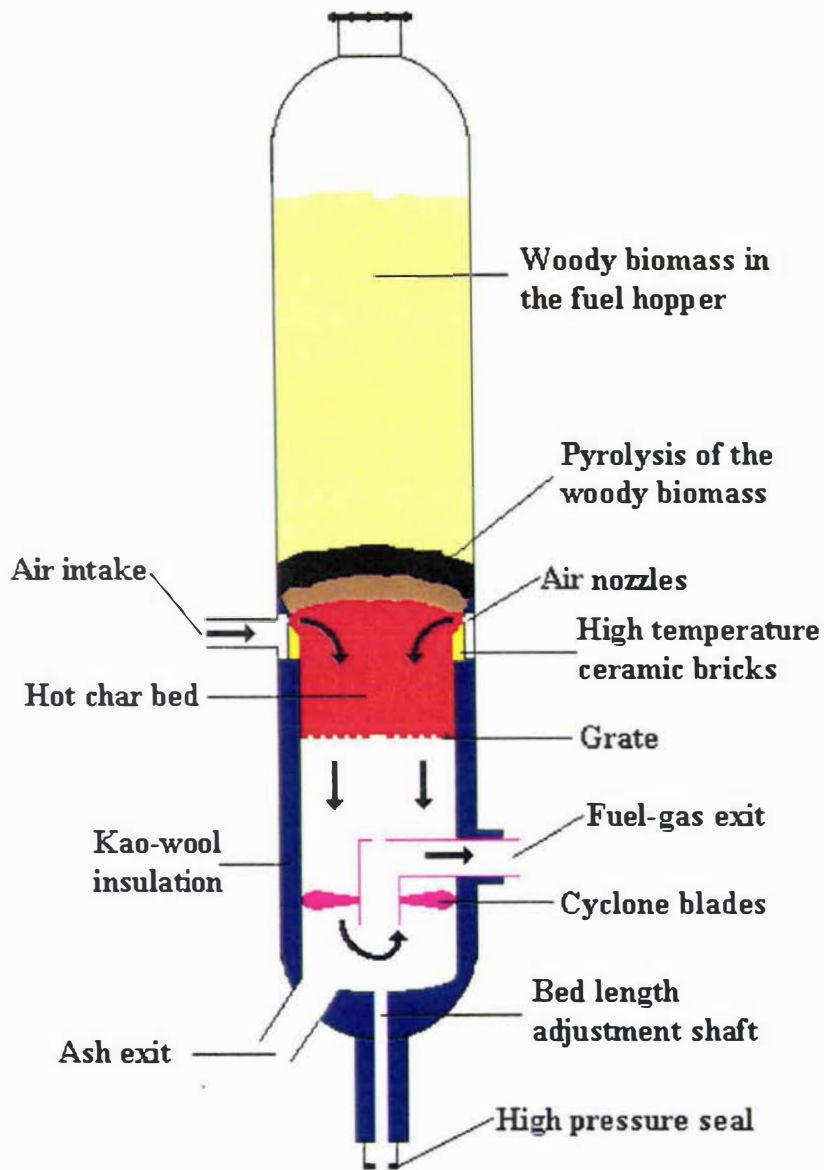
Three problem areas had been identified when operating the gasifier. Firstly, the fuel above the bed was occasionally prone to bridging. Bridging of the fuel above the bed prevented the even flow of fuel into the bed. While it is possible to cope with this problem using woodchip, it was considered improbable that stringy fuels such as bark mulch could be used. Secondly, the pressure transducers would resonate under certain conditions. This made data collection difficult. Thirdly, in exploratory experimental work it was found that at longer bed lengths the gasifier was prone to blocking. Examination of the bed indicated a build up of fine particulate material above the grate. This negative behaviour interfered with planned experiments, therefore it was necessary to determine the conditions under which the blocking became a problem.

4.2 Method

4.2.1 Widening of the gasifier

The most obvious way of removing the bridging problem was to increase the diameter of the gasifier hopper feed and bed throat (Figure 4.1). The diameter of the hopper feed was increased from 160 mm to 240 mm and the bed throat (originally 120 mm) was removed. The diameter just above the grate remained 200 mm.

Figure 4.1 Modified gasifier



4.2.2 Filtering noise

The first task was to identify the frequency range of the observed resonance phenomena. This was done by rapidly decreasing the pressure differential over

the transducer and monitoring the voltage response using the oscilloscope function of the logger. The pressure differential was obtained by sucking on the low pressure side of the transducer. A rapid pressure differential change was achieved when the suction was released again.

4.2.3 Compaction of the gasifier bed

The gasifier was run with an air flow rate of $0.025 \text{ m}^3.\text{s}^{-1}$ at a pressure of 670 kPa. Prior to experimental work the gasifier was given a warm up period. For each data point the bed length was manually adjusted and the gasifier was reloaded with wood-chip (4% m.c.). During the experiment the pressure drop transducer was broken and replaced.

The following parameters were monitored in real time using a supreme mini-pod data logger:

- gasifier pressure
- volume flow rate of air into the gasifier
- input air temperature
- pressure drop through the bed
- fuel-gas exit temperature
- frictional function
- molar flow rate of air into the gasifier

4.3 Results and Discussion

4.3.1 Widening of the gasifier

The gasifier hopper feed and bed were widened in an attempt to prevent bridging (Figure 4.1). To date, no bridging has been observed, even when using stringy fuels such as bark mulch.

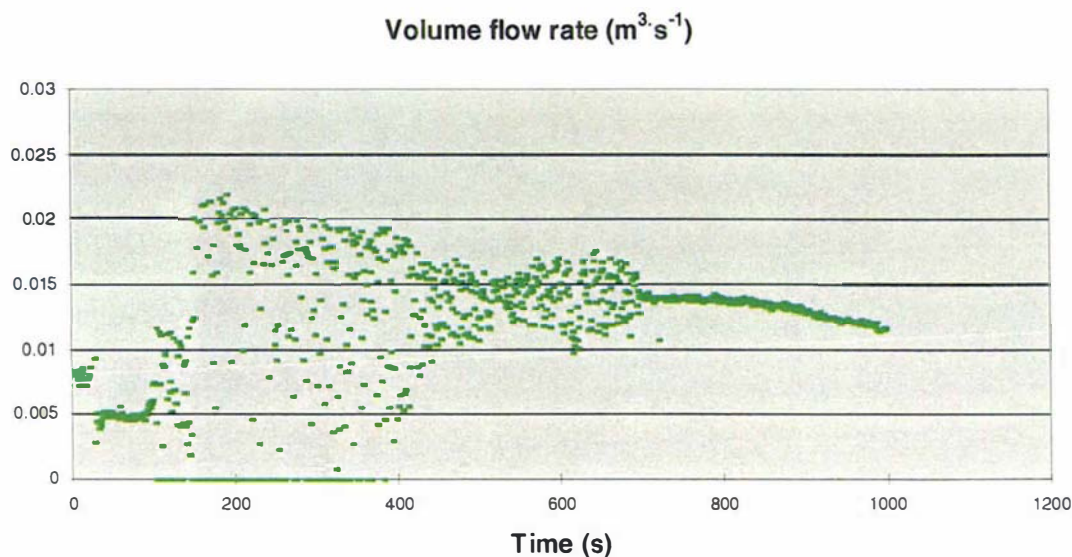
In addition the following extra modifications have been made:

- The mild steel cyclone was deteriorating from temperature induced flaking. This was replaced with a high temperature stainless steel (grade 253MA) cyclone.
- The cast iron steam valve used to control the exit fuel-gas flow was prone to insert seat failure. The insert seat would lift out, shutting off the gas flow. The cast iron steam valve was replaced by a purpose made stainless steel valve (grade 316).
- The thin sheath on the thermocouple was subject to ash induced corrosion. The thermocouple was enclosed in an additional stainless steel sheath.

4.3.2 Filtering transducer noise

It was found that the pressure transducers used to measure the volume flow rate and the bed pressure drop exhibited resonant behaviour. An example of this phenomenon is shown in Figure 4.2. Notice that the resonant behaviour can also stop (700 seconds) without any changes being made to the gasifier.

Figure 4.2 Transducer noise



By doing rapid pressure drop experiments it was found that the transducers had a natural resonant frequency. In Figure 4.3 the pressure is rapidly dropped by releasing suction on the low pressure side of the transducer. Notice that the unfiltered line (red) resonates. Fourier transform of the unfiltered response showed a resonant frequency of about 20 Hz (Figure 4.4). The gasifier is audibly very noisy. Therefore it is possible that internal 20 Hz noise from the gasifier caused the transducers to resonate.

Two methods were devised to combat the noise problem:

1. Electronically filter all signals below 1 Hz. Figure 4.3 shows both transducers undergoing a pressure drop. One of the transducers (blue line) has been electronically filtered.
2. Physically filter the signal by inserting a small orifice in the pressure lines; thus, slowing down the response time of the transducer. This was achieved by inserting a no. 16 syringe needle. Figure 4.5 shows both transducers undergoing a pressure drop. One of the transducers (blue line) has been filtered using a syringe needle.

A combination of electronic and physical filtering was subsequently used for the volume flow rate transducer. Only electronic filtering was used on the pressure drop transducer, because tar from the fuel-hopper had a tendency block the orifice of the needle.

Figure 4.3

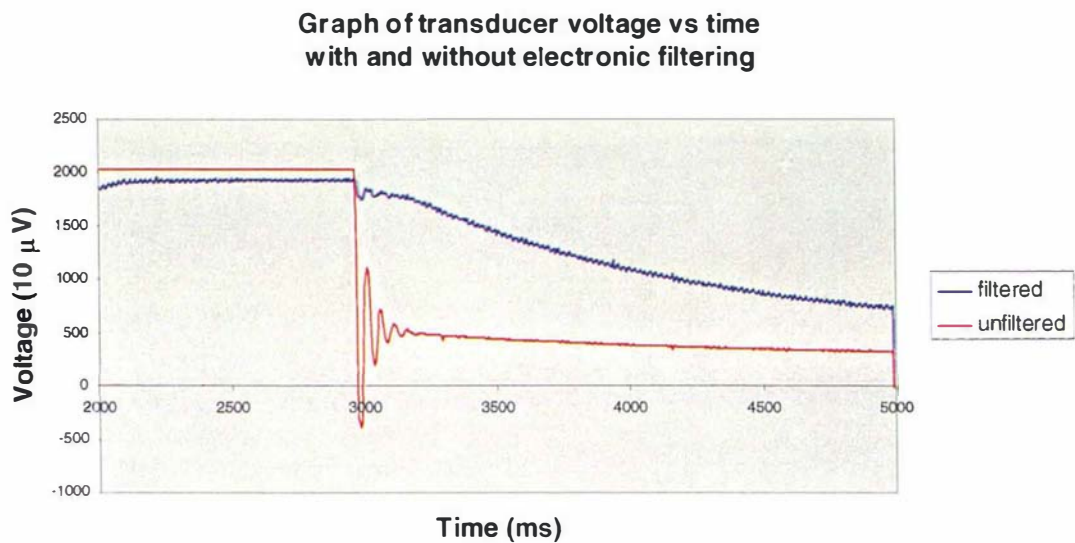


Figure 4.4

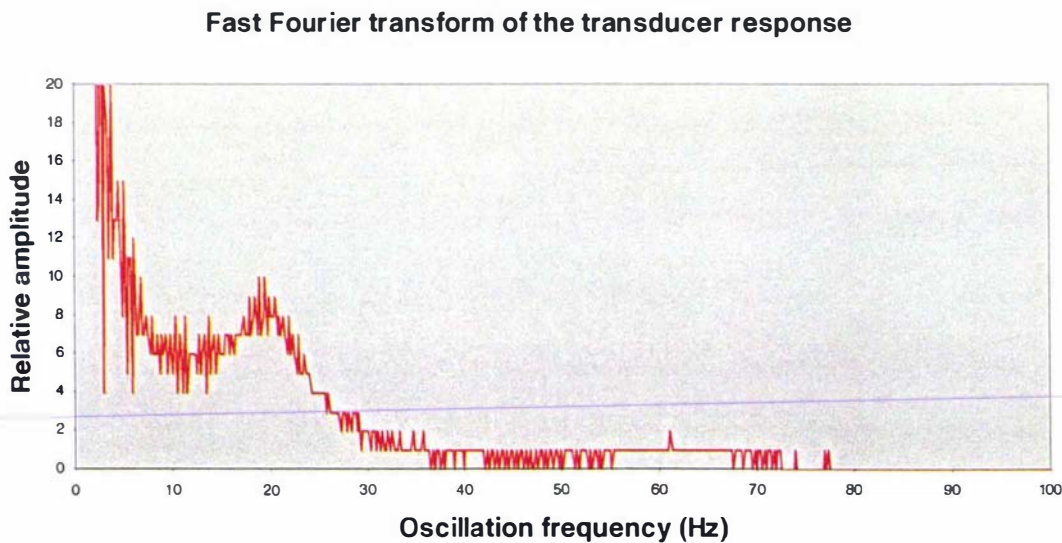
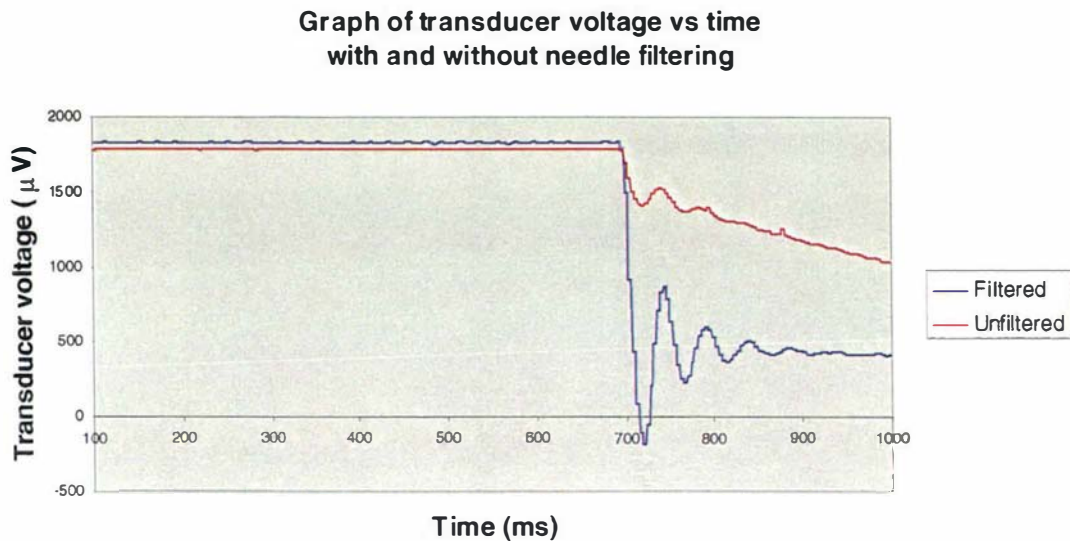


Figure 4.5



4.3.3 Compaction of the gasifier bed

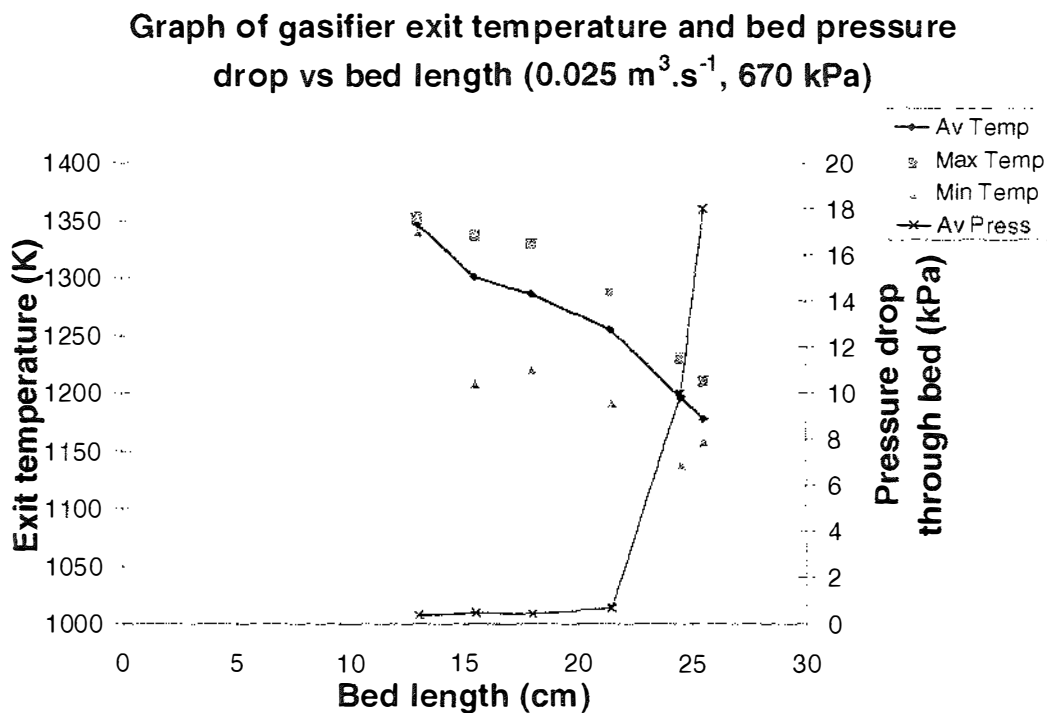
The traditional downdraft gasifier has an excess of char in the bed. This excess component is sometimes called a dead zone, because there is insufficient thermal energy remaining to allow further gasification (Reed, 1981). In this zone there is almost no chemical removal of char. Because there is no chemical removal, fine particulates build up in the dead zone. This build up can result in blockage of the gasifier.

The traditional method of overcoming blockage is to physically remove the char from the bottom (dead zone) of the bed. Often these systems are in the form of a moving grate. Aside from the added complication, a disadvantage of this method is that the chemical energy of the char is removed from the gasifier also. Typically a moving grate removes 5 - 10 % of the available chemical energy from the system in the form of char (Reed, 1998).

At exit temperatures less than 1200 K, high pressure drops occurred as the gasifier became blocked with fine particulate materials (Figure 4.6). Note that the recorded pressure drop at 24 cm bed length was still increasing at the end of the run.

At exit temperatures 1250 - 1300 K, the gasifier does not block. The observed pressure drops are very low. The exit temperatures in this range vary considerably; note the maximum and minimum values also shown in Figure 4.6.

Figure 4.6



At short bed lengths a maximum exit temperature was reached at 1350 K. When the downdraft gasifier was run at the upper temperature limit, very stable exit temperatures were obtained as indicated in Figure 4.6. Initially this was considered to be a region of exit temperature stability. This was later found to be a fault in the logger hardware. The thermocouple voltage was within the range

of the logger, but temperatures beyond 1350 K were not recorded properly. This resulted in loss of data.

4.3.4 Pressure drop mathematical development

When examining the blockage problem there are two key issues to consider. Primarily there is the problem build up of char in the bed, which may be attributed to a low chemical removal rate of char from the bed. Secondly there may be a pressure drop limit, which if exceeded, could cause compaction and blockage of the bed. The following is a continuation of the mathematical development started in chapter 2.

$$F_D = \frac{A_{ss} \rho_{ms} u^2 k_z}{g_c} \quad (2)$$

The pressure drop through the section is equal to the drag force on the section divided by the cross-sectional area of the bed.

$$\Delta p_s = \frac{F_D}{A_{cs}} \quad (32)$$

where

A_{cb} = cross-sectional area of the bed (m^2)

Δp_s = pressure drop through the section (Pa)

Substituting (2) into (32)

$$\Delta p_s = \frac{A_{ss} \rho_{ms} u^2 k_2}{A_{cb} g_c} \quad (33)$$

The velocity is equal to the volume flow rate in the section ($v_s, m^3.s^{-1}$) divided by the cross-sectional area of the channels in the section (A_{cs}, m^2).

$$u = \frac{v_s}{A_{cs}} \quad (5)$$

Substituting (5) in (33)

$$\Delta p_s = \frac{A_{ss} \rho_{ms} v_s^2 k_2}{A_{cb} A_{cs}^2 g_c} \quad (34)$$

From equation (15)

$$v_s = \frac{f_i R T_s X_s}{p_i} \quad (15)$$

Substituting (15) into (34)

$$\Delta p_s = \frac{A_{ss} \rho_{ms} f_i^2 R^2 T_s^2 X_s^2 k_2}{A_{cb} A_{cs}^2 g_c p_i^2} \quad (35)$$

From equation (23)

$$\rho_{ms} = \frac{p_i Y_s M_{r_{air}}}{R T_s} \quad (23)$$

Substituting (23) into (35)

$$\Delta p_s = \frac{A_{ss} f_i^2 R T_s X_s^2 Y_s M_{r_{air}} k_2}{A_{cb} A_{cs}^2 g_c p_i} \quad (36)$$

The pressure drop through the bed is equal to the sum of all the pressure drops through the sections.

$$\Delta p_b = \sum_{s=1}^n \Delta p_s \quad (37)$$

Δp_b = pressure drop through the bed (Pa)

Substituting (36) into (37)

$$\Delta p_b = \sum_{s=1}^n \frac{A_{ss} f_i^2 R T_s X_s^2 Y_s M_{r_{air}} k_2}{A_{cb} A_{cs}^2 g_c p_i}$$

Simplifying

$$\Delta p_b = \frac{f_i^2 R M_{r_{air}} k_2}{A_{cb} g_c p_i} \sum_{s=1}^n \frac{A_{ss} T_s X_s^2 Y_s}{A_{cs}^2} \quad (38)$$

An objective of this technology is to get larger molar flow rates through the downdraft gasifier. If pressure drop becomes a limiting feature, then from (38) pressure drop can be reduced in three ways:

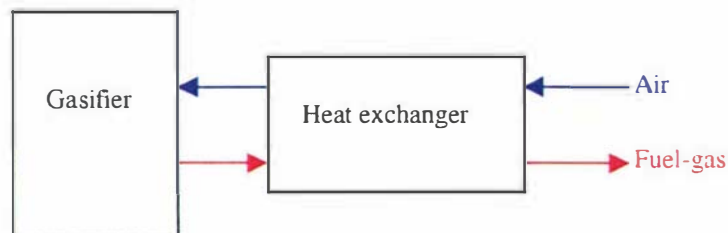
1. Increase the gasifier pressure.
2. Reduce the surface area of the char particles in the bed by decreasing the bed length
3. Change the particle size distribution in the bed ($\frac{A_{ss}}{A_{cs}^2}$). The finer the particles

the greater the surface area. This is achieved by increasing the exit temperature, which removes the fine components from the bed. i.e. reduce the proportion of fine particulates in the bed. This also fits with experimental results in Chapter 3 where lower than expected compression energy losses were observed at elevated exit temperatures.

4.3.5 Increasing the chemical energy content of the fuel-gas

A normal atmospheric pressure downdraft gasifier reaches an optimum conversion of wood energy into fuel-gas energy when the exit temperature is 1025 K (Desrosiers, 1981). By running the gasifier with a higher exit temperature the fuel-gas chemical energy content will decrease. To compensate for this decrease, the intake temperature can be increased. This can be accomplished in a number of ways, two such methods are as follows:

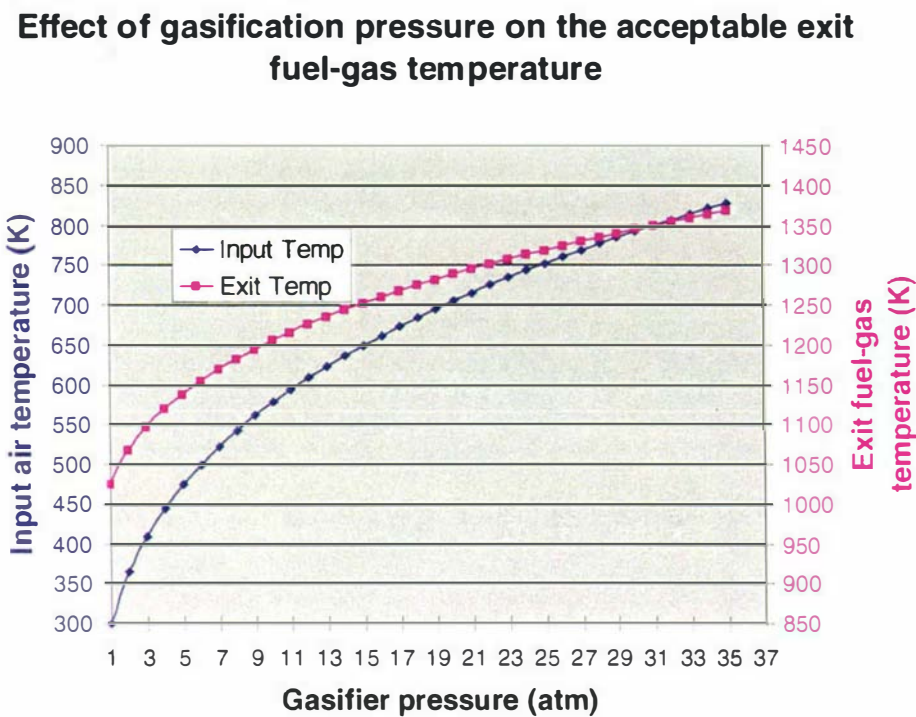
1. Use heat from the exit gas stream to pre-heat the intake gas stream. A similar process is used in low pressure directly heated IGCC systems (Craig & Mann, 1997).



2. In a high pressure gasifier, adiabatic heating of the input air occurs from compression to high pressure (no intercooler). From Figure 4.8 it can be seen that at pressures of 2500 kPa, an exit temperature of 1320 K has the same chemical energy as an atmospheric gasifier operating at 1025 K. This was calculated by assuming a heat ratio of 1.54.

Either method or a combination of methods will allow gasification at higher exit temperatures while still obtaining good conversion to fuel-gas chemical energy.

Figure 4.8



The technology being explored utilises elevated temperature to achieve higher reaction rates. Much of the focus has been upon increasing surface kinetic reaction rates. The surface kinetic activation energy for char is 183 kJ.mol⁻¹ (Desrosiers, 1981). Using the Arrhenius equation, a graph of the relative surface kinetic reaction rate vs temperature can be constructed (Figure 4.9). A pre-exponential factor of 2.119x10⁹ was used to obtain a relative rate of 1 at 1025 K.

$$k = Ae^{\frac{-E_a}{RT}}$$

Arrhenius equation

Where

- k = Reaction rate
- A = Pre-exponential factor
- E_a = Activation energy (J.mol⁻¹)

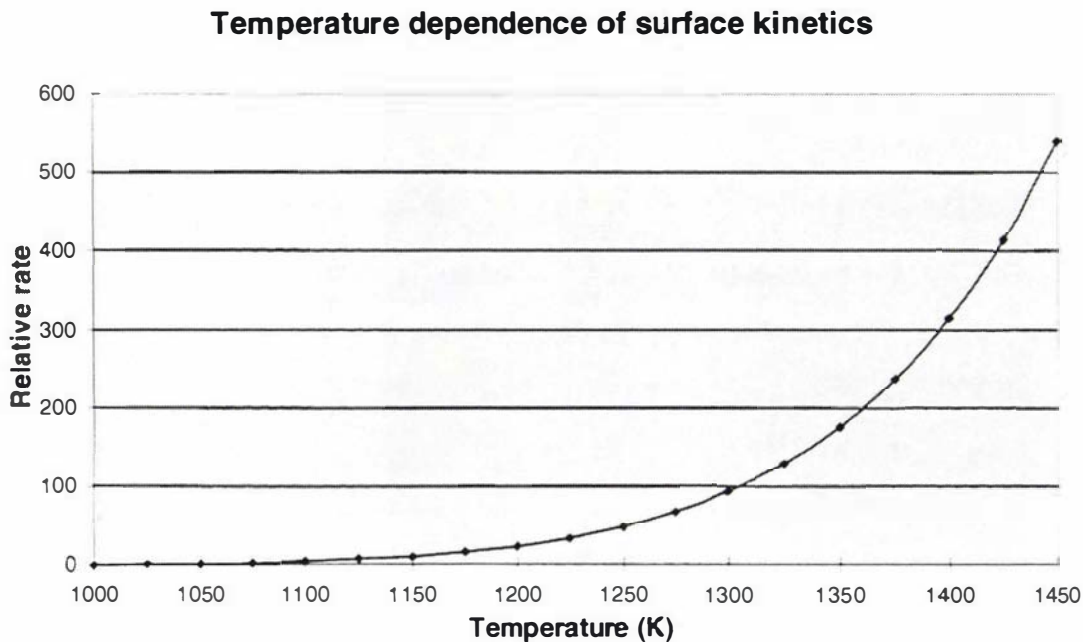
R = Gas constant ($8.314 \text{ J.K}^{-1}.\text{mol}^{-1}$)

T = Temperature (K)

There are practical materials limitations that will influence the maximum exit temperature of the gasifier. Entrained flow coal gasifiers have gasifier exit temperatures of up to 1775 K (Kurkela *et. al.*, 1993). Some biomass updraft designs have grate temperatures of 1675 K (Reed, 1998). The grate and cyclone of the gasifier used in this study are constructed from high temperature stainless steel (253MA). The maximum continuous operating temperature of this grade of stainless steel is 1425 K. Therefore using the current materials technology the maximum continuous exit temperature would be 1425 K.

From Figure 4.9 it can be seen that the surface kinetic reaction rate at 1425 K is over 400 times greater than at 1025 K. Thus, at 1425 K the rate of chemical removal of char particulates from the bed can be much greater than at 1025 K. Therefore the requirement for physical removal of the char from the bed is reduced. This feature will be a great asset when utilising fuels like bark mulch, which form fine char particulates.

Figure 4.9 Calculated using a surface kinetic activation energy of 183 kJ.mol⁻¹ (Desrosiers, 1981). The relative reaction rate at 1025 K has been designated a value of 1.



4.4 Conclusion

Attempts to solve bridging, blocking and transducer operational problems were successful. It was postulated that bed pressure drop may be a limiting factor in gasifier throughput. The mathematical model was advanced to provide a theoretical description of the bed in terms of pressure drop.

For the following reasons future experiments should be designed to run at a high exit temperature:

- High rate of chemical removal - No need for physical removal of char from the bed
- The condition is achieved at short bed lengths with low pressure drops

- High volume flow rates can be used
- The average surface kinetic reaction rate is greatly increased
- Simplifies problems of using difficult fuels

At high exit temperatures, the decreased chemical energy content of the gas stream can be compensated for, by increasing the intake air temperature.

5.0 Gasifier scale-up

5.1 Introduction

This PhD project has a commercial bias. An objective of the project was to provide information to assess the potential for this technology in a commercial energy plant. The following criteria add to viability of an energy system:

1. The energy application should preferably be co-generation. Co-generation plants are common in the existing forestry industry. They have the advantage that both heat and electricity are valuable.
2. The energy system should be run continuously. Continuous running reduces the capital contribution of the energy cost.
3. The energy system should be big enough that economy of scale is achieved. This has the advantages of higher electrical efficiency, lower labour costs, and lower capital costs per kilowatt. Basic analysis by Steve Goldthorpe (presentation) has shown that co-generation plants at the 50 MW_t scale are a likely choice. This is the approximate size of the bark-mulch component of the combustion system used at the Kinleith pulp mill.

Pressure is a key component to the scaling up of the downdraft gasifier. High pressure IGCC systems have been modelled (Graig and Mann, 1997) at pressures of 3.24 MPa (32 atm) The air pressure supply at the experimental site is a shared facility. The maximum pressure generally available at the site is 700 kPa. Therefore data is collected at these lower pressures (100-700 kPa) and extrapolated to higher pressures.

5.2 Method

The gasifier was run with an air flow rate of $0.025 \text{ m}^3 \cdot \text{s}^{-1}$ at pressures of 110, 220, 330, 450 and 670 kPa. Prior to experimental work the gasifier was given a warm up period. For each data point the bed length was manually adjusted and the gasifier was reloaded with dry wood-chip (4 % m.c.). The following parameters were monitored in real time using a supreme mini-pod data logger:

- gasifier pressure
- volume flow rate of air into the gasifier
- input air temperature
- pressure drop through the bed
- fuel-gas exit temperature
- molar flow rate of air into the gasifier

5.3 Results and Discussion

5.3.1 Pressure effects

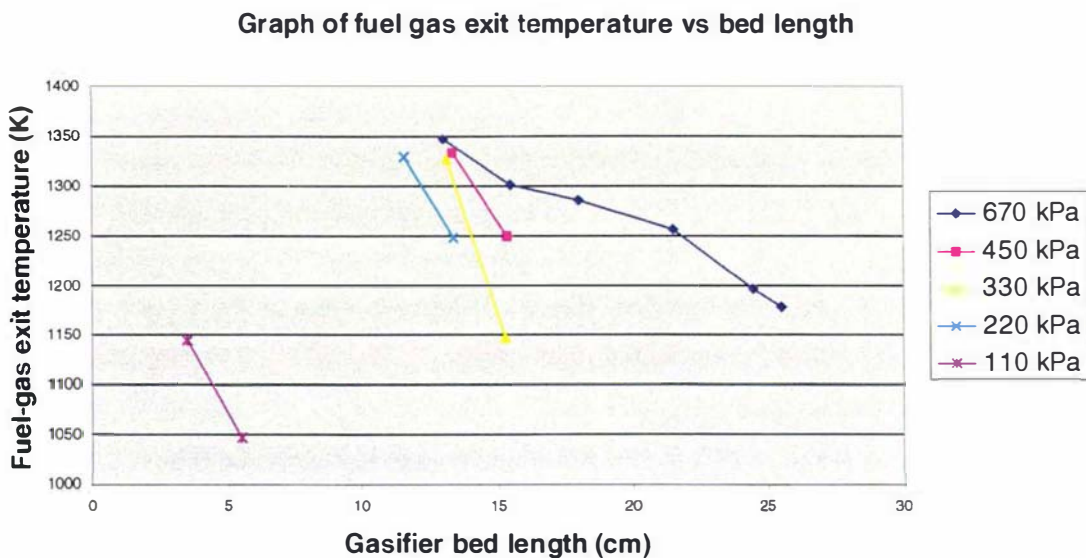
A graph of exit temperature versus bed length over a range of gasifier pressures is shown in Figure 5.1. Part of the original data from this set was left out due to a logger hardware fault. Running con-currently with the discovery of data loss, were plans to upgrade the gasifier.

The new gasifier would be capable of longer gasifier runs and would produce better data. Furthermore, the new gasifier would comply with the draft version of the new pressure vessel regulations. Therefore it was decided by ECNZ and

Massey that the thesis should be written based on the data available. Any continuation of the pressure experiments would be done with the new gasifier. It is also hoped that experimental data might be obtained at higher pressures, though this will depend on the availability of a suitable air supply.

Some of the data at 1350 K is shown in Figure 5.1. By including the first of these data points information on the general trend of the data can be appreciated, but it must be remembered though that the true exit temperature at these points is greater than 1350 K.

Figure 5.1



5.3.2 The correctly operating downdraft gasifier

In a correctly operating downdraft gasifier, gasification of the pyrolysis gases is very rapid relative to gasification of the char. It is desirable that the gasification

of the pyrolysis gases goes to completion while there is still excess energy (temperature) available to do so. If insufficient energy (temperature) remains to gasify the pyrolysis gases, some pyrolysis gases may be carried through as tar.

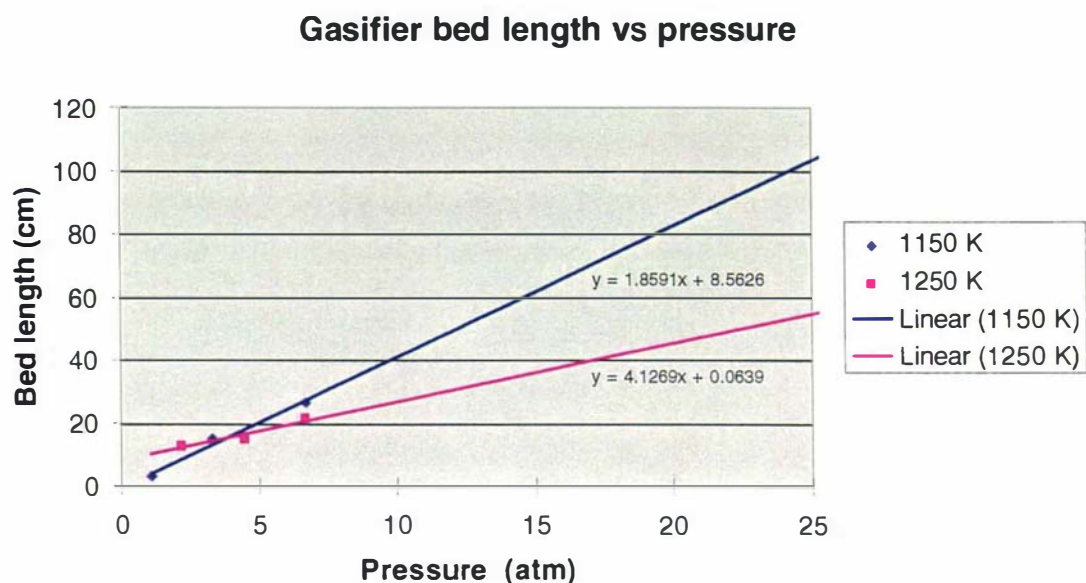
5.3.3 Gasifier reactions

Low temperatures char reactions

At low temperature gasification reactions for the char are slow. There is always a saturation of reactive gases about the char reactive sites. The reaction kinetics is said to be dependant on surface kinetics (Graboski, 1981). At low temperatures these reactions have a reaction order less than first order with respect to pressure. This means that the reaction rate does not increase in proportion with the pressure. To obtain greater throughput under these conditions a greater volume of reactive sites is needed. Increasing the bed length increases the volume of active char sites.

From Figure 5.2 it can be seen that at low temperatures (1150 K) the required bed volume dramatically increases with increasing pressure. Note that the slope of the line at 1150 K is less than the slope of the line at 1250 K indicating a movement toward zero order at low temperature. If the reaction order were zero with respect to pressure, then the required bed volume would be proportional to the pressure.

Figure 5.2 is a replot of extrapolated data from Figure 5.1



High temperature char reactions

At high temperature char surface kinetics are rapid. Gasification reactions at high temperature depend on the mass flow rate of gases to the char reactive sites (Graboski, 1981). From figure 5.1 and 5.2 it can be seen that at high exit temperatures the required bed volume is less dependant on the pressure indicating a movement toward first order reactivity.

Pyrolysis gas reactions

Pyrolysis gas reactions are similar to char reactions, but with a significant difference. Unlike char, pyrolysis gas is compressible. Hence at low temperature the reactions are at least first order with respect to pressure. At high temperatures the reactions approach second order.

5.3.4 The effect of pressure on tar cracking

At all temperatures pyrolysis gas reactions have a greater reaction order with respect to pressure than do char reactions (because pyrolysis gas is compressible, char is not). Therefore, increasing the pressure further increases the difference in relative reaction rate of pyrolysis gas and char. This aids tar cracking.

Tar cracking improves with both the partial pressure of tar cracking gases (Section 1.7) and the temperature. A simple way to describe the effect of pressure on tar cracking is to consider an example. If the downdraft gasifier were operated at two pressures (670 kPa and 110 kPa) with a bed length 15 cm and a volume flow rate of 25 l.s^{-1} , then the expected exit temperature for the high pressure run would be about 1300 K. The low pressure case would be $<1050 \text{ K}$ (Figure 5.1). In both cases the gas residence time (reaction time) in bed is identical. The high pressure case would have a higher average temperature and a higher partial pressure of tar cracking gases.

These arguments strongly suggest that the tar cracking ability of the downdraft gasifier will be improved at higher pressures. The traditional downdraft gasifier produces a gas with low tar content. The HTP downdraft will be even better. Experimental measurements of the fuel-gas tar content are planned for next year.

5.3.6 HTP scale-up of the downdraft

One of the objectives of this research project is to obtain information that will aid the design of higher output gasifiers. Evidence collected to date, indicates that the output of the downdraft gasifier is increased with high exit temperature and high pressure (HTP) in two ways:

1. Increased molar flow of gas through the gasifier.
2. Chemical changes which allow larger gasifier dimensions without compromising tar cracking ability.

Ideally the thermal output would be experimentally determined at High Pressure IGCC/IGAC gasification pressures. However three factors prevent this in the current PhD project:

1. An air supply is not available.
2. The current fuel capacity of the gasifier is insufficient for high output runs.
3. The gasifier has not been designed for these pressures.

By extrapolating data obtained at pressures up to 670 kPa, it is possible to obtain a crude estimate of the thermal output of the HTP gasifier at 1500-2500 kPa (Table 5.1). The bed length was calculated by extrapolation of data in Figure 5.2. In doing so an attempt is made to account for kinetic behaviour. The bed length required to reduce the exit temperature to 1250 K increases with the pressure (and molar flow) of air into the gasifier. Changes in the bed length affect the surface of char particles in the bed.

The pressure drop was calculated by assuming equation (38). For the sake of calculation simplicity it was assumed that the bed length was proportional to the surface area of char particles. None of the calculated bed pressure drops exceeded the maximum experimental observed stable pressure drop (8 kPa). The

thermal output was calculated from gas chromatography data obtained in chapter 6.0.

A valid criticism of these calculations is that data is being extrapolated a long way given the inherent uncertainty of factors such as particle distribution. However, the crude estimates err on the side of being conservative for the following reasons:

- The gas chromatography data used in the calculations was obtained at a bed length of 10 cm. The exit temperature was at least 1350 K (i.e. not 1250 K). Hence the chemical energy content used in the calculations was artificially low.
- The calculations assume no pre-heating of the intake air. However in the real situation the intake air would be preheated, thus increasing the chemical energy of the gas stream.
- The calculations were based on data using a volume flow rate of about 25 l.s⁻¹. This volume flow rate was used for the experimental because of the limited fuel capacity of the hopper. It is possible to increase the volume flow rate.
- The modelling was done at 1250 K because data were available at this exit temperature. However, greater throughput would be possible at shorter bed lengths. Short bed lengths have lower pressure drop and lower bed length pressure dependence (Figure 5.1). Therefore there is even greater scope to increase the volume flow rate.

Table 5.1 Crude estimates of gasifier throughput at gas turbine pressures

Pressure (kPa)	Molar flow rate of air (mol.s ⁻¹)	Extrapolated bed length (cm)	Calculated pressure drop through the bed (kPa)	Calculated thermal throughput per bed cross-sectional area (MW _t .m ⁻²)
2500	24.6	55.0	6.6	105
2000	19.7	36.4	4.4	84
1500	14.7	27.2	2.6	63

It is claimed that many groups operate an atmospheric pressure stratified downdraft gasifier with a thermal throughput of 3.2 MW_t.m⁻² (Reed *et. al.*, 8-31, 1987), but that this can climb to at least 6.5 MW_t.m⁻² with technologies such as automated char removal systems and oxygen. The figure of 6.5 MW_t.m⁻² is recommended for the sizing of an air atmospheric pressure downdraft gasifier (Reed *et. al.*, 8-31, 1987), although other claims in the same document (4-16) suggest a throughput of 9.5 MW-lhv.m⁻² is possible.

The calculations predict thermal throughputs greater than 105 MW_t.m⁻² might be possible at pressures of 2500 kPa. No serious attempt was made during this PhD study to experimentally determine the maximum throughput. Instead the volume flow rates used in the gas chromatography measurements (Chapter 6) were focussed on fuel conservation.

The highest molar flow rate of air into the gasifier (10.2 mol.s⁻¹) was used in the pre-steady state experimental work. A throughput of 43 MW_t.m⁻² was calculated, based on the moisture content of the fuel and the expected fuel-gas energy content. Similarly for the bark-mulch experiment (Table 6.2) a throughput of 37 MW_t.m⁻² is predicted. The maximum thermal throughput for which the gas analysis is known, rather than assumed, was 29 MW_t.m⁻². This was obtained in the fuel flexibility moisture content experiments.

Future research should be focussed on the experimental (not extrapolated) demonstration of HTP downdraft gasifier technology at IGCC/IGAC pressures. For this to happen, a suitable air supply source must be obtained. This does not appear to be likely from within New Zealand. Many countries have high pressure IGCC research programs and are equipped with a suitable air supply. Of these countries, Australia is geographically closest.

5.3.7 Dimensional scale-up of the downdraft gasifier

In addition to scaling up with HTP there are also dimensional methods of scaling up. There are two common downdraft gasifier configurations. In one method air is introduced from the periphery of the gasifier bed Figure 5.3 (Kurkela, 1993). The other method is to introduce air from the centre of the gasifier bed Figure 5.4.

In both methods the tar generated above the air nozzles passes through the bed at the zone of lowest pressure. This occurs furthest from the air nozzles. It is important that the temperature in this low pressure zone is high enough that all of the tars crack when passing through. If the bed diameter is too large, the low pressure zone may allow tar to pass through the bed uncracked.

By combining these two methods (Figures 5.3 & 5.4), air can be introduced from both the periphery and the centre Figure 5.5. In doing so it is possible to increase the cross-sectional area of the bed. Another alternative is to elongate the bed into a trough shape. The trough beds can be stacked horizontally in a grid and are simple to manufacture.

Figure 5.3 Air introduced from bed periphery.

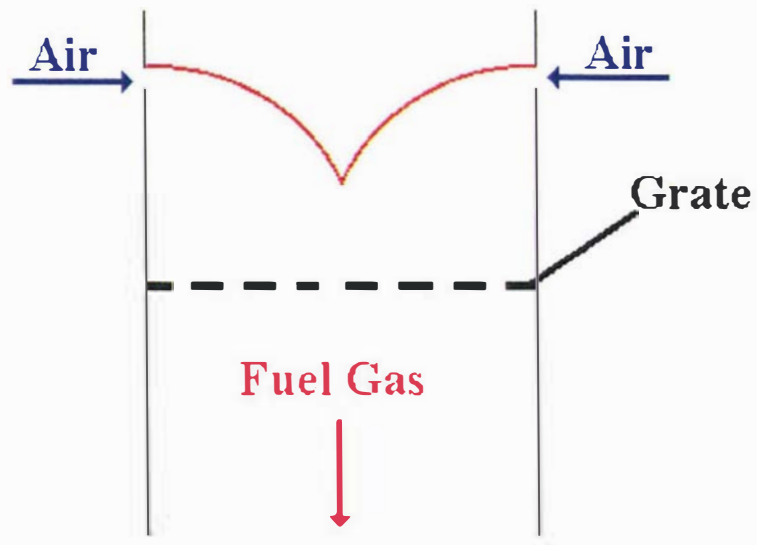


Figure 5.4 Air introduced from the centre

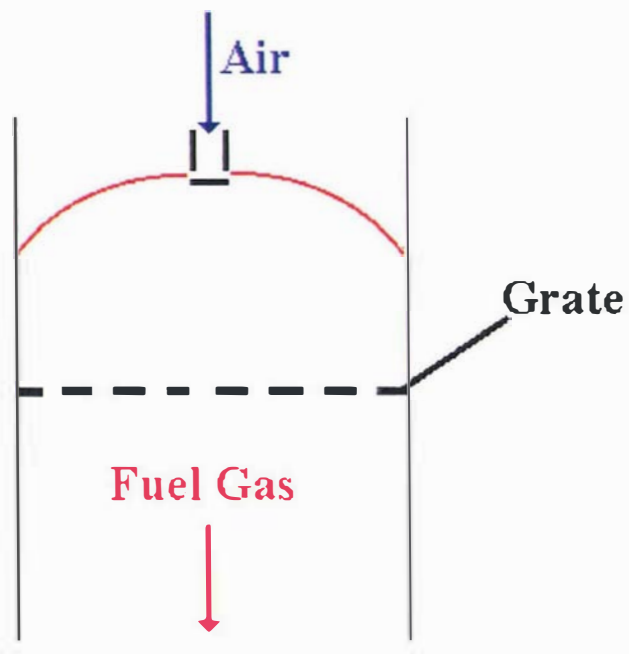
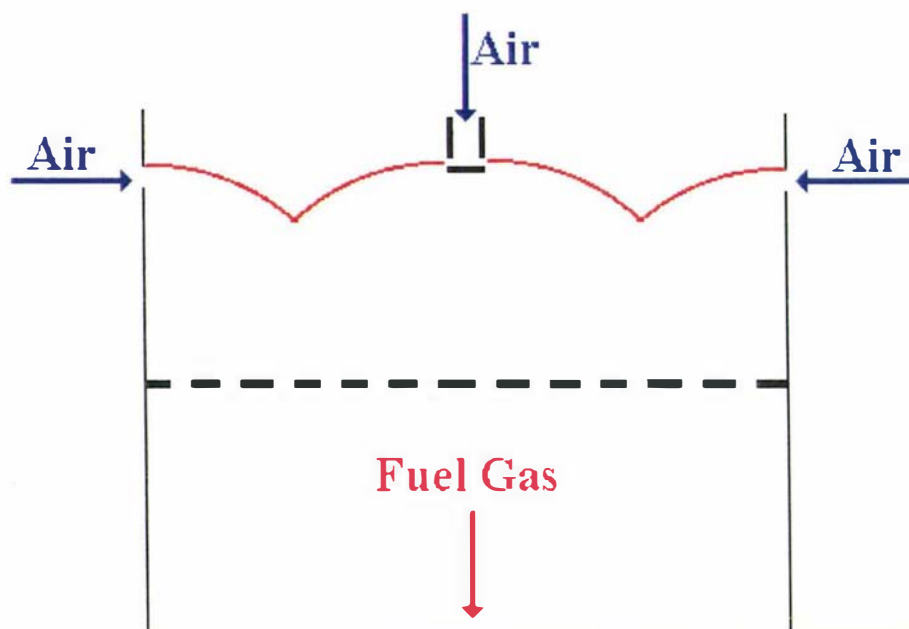


Figure 5.5 Air introduced from the periphery and the centre.



In Table 5.2 estimates have been made of the cost of a 400 mm bed diameter HTP downdraft gasifier. The thermal output at 2500 kPa is expected to be at least 13 MW_t. An automated fuel feed system has not been included. No allowance has been made for specialised start up technology needed for wet fuels.

The estimated HTP downdraft cost per kilowatt of capacity is only N.Z. \$1.2/kW_t. A high pressure PDU gasifier including feeding system (156 MW_t) was costed at U.S. \$27.25 M (1997) (Craig and Mann, 1997). Using the current exchange rate of 0.52, this is equal to N.Z. \$340/kW_t. Clearly the capital costs for the HTP downdraft are very low. Even if the throughput were halved and the capital costs of the gasifier increased 10 fold the HTP downdraft would still be a very inexpensive option.

So why are fluidised bed gasifiers so expensive? Consider the example of the 156 MW_t PDU gasifier. The fluidised bed inside diameter is 2.86 m (i.e. 6.42 m² cross-sectional area) (Craig and Mann, 1997). The thermal output per square

meter is $24 \text{ MW}_t \cdot \text{m}^{-2}$. The diameter of the disengaging zone isn't given, but the cross sectional area of the disengaging zone is normally at least twice the area of the bed.

The fluidised bed gasifier has a thick refractory lining. The total gasifier diameter including the refractory lining is 4.26 m (i.e. 14.25 m^2 cross-sectional area). If the thick refractory lining is included, then the thermal throughput is effectively only $10.9 \text{ MW}_t \cdot \text{m}^{-2}$. The fluidised bed gasifier does not have an especially high throughput per cross-sectional area and the total bed length is much longer than the HTP downdraft gasifier.

Table 5.2

Basic Gasifier for IGCC	Cost (NZ \$)
Hopper (1000 litre)	\$10000
Gasifier Shell	\$3000
Internal Components	\$1750
Support Structure	\$1000
Subtotal	\$15750
Additional costs for a stand alone experimental gasifier	
Internal cyclone blade	\$600
Char Ash Collector	\$300
Exit Valve	\$500
Analysis Transducers	\$1500
Total	\$18650

A 50 MW_t bio-energy plant may use a combination of a number of smaller HTP downdrafts (e.g. $5 \times 13 \text{ MW}_t$), or a single large gasifier. An advantage of smaller gasifiers is that they may be cheaper to manufacture and transport.

Combinations of small gasifiers are more flexible to cope with variations in plant size. Furthermore, by having a slight capacity excess, the gasifiers can be serviced in rotation without the whole system being shut down. By contrast, a single large gasifier takes up less space and has less complicated piping. The optimum size has yet to be determined.

5.4 Conclusion

Ballpark estimates of the throughput at gas turbine pressures were obtained by extrapolation of the data. These predicted throughputs much greater than the standard atmospheric downdraft. Pre-heating of the intake air, shorter bed lengths and higher molar flow rates will further contribute to the throughput. An experimental program at pressures up to 2500 kPa is recommended.

High temperature and pressure aid tar cracking. Therefore the HTP downdraft is expected to out perform the tar cracking ability of the tradition atmospheric downdraft gasifier.

The HTP downdraft gasifier capital costs are very low. Even if the throughput were halved and costs were severely underestimated, the HTP downdraft would still be inexpensive.

6.0 Fuel flexibility

6.1 Introduction

The pulp industry is currently the largest producer of electricity from woody biomass. Therefore the pulp industry is a very important market for IGCC technology. To a lesser extent, woody bio-energy is also used in large timber mills for electricity production and timber drying.

Electricity production from bio-energy plantations is unlikely to be viable in New Zealand. Growing biomass to burn is not a commercially competitive use of land. New Zealand does not have a policy of subsidising renewable energy, and this is unlikely to change in the foreseeable future. Woody biomass from land based effluent treatment is a possibility, but these systems have yet to be established. High capital, transport and harvesting costs are severe limitations. Hence this technology may not be commercially viable.

Pulp and paper production is very energy intensive, requiring both heat and electricity. Often pulp companies do not produce enough cheap waste biomass to meet their energy requirements. Therefore other energy sources are recruited like electricity, geothermal heat, natural gas and coal. Table 6.1 shows energy sources used by the U.S. pulp and paper industry (American Forest & Paper Association, 1997).

Table 6.1

U.S. pulp and paper					
Energy Source: Self-Generated	1972	1994	Energy Source: Purchased	1972	1994
Spent pulping liquor	33.0	40.8	Residual fuel oil	21.2	6.0
Bark	4.5	6.6	Distillate fuel oil	1.0	0.3
Wood residues	2.0	7.3	Natural gas	21.1	16.9
Self-generated hydropower	0.4	0.2	Purchased electricity	4.4	6.4
Other	0.4	1.0	Coal	10.7	12.5
			Other	1.3	2.0
TOTALS	40.3%	55.9%		59.7%	44.1%
Note: The percentages for each source represent the fraction of total energy used by the pulp and paper industry in the year for which data are reported.					

The United States is the largest consumer of pulp and paper products in the world. The U.S. pulp and paper industry, which supplies the bulk of this demand, is larger than the next four largest producers of pulp and paper outside the United States combined (USAEP, 1997).

From Table 6.1 it can be seen that more energy is derived from fossil fuels than bark and wood residues. A local example of this is the Carter-Holt Harvey Kinleith mill, which has just installed a 34 MWe biomass/natural gas co-fired power generation system. This combustor obtains 50% of its energy from natural gas. Furthermore co-combustion with coal is also being investigated.

Agenda 2020 outlines the research objectives for the United States American Forest and Paper Association (AF&PA, 1997), as agreed on by industry representatives and related trade and research organisations with support from the U.S. Department of Energy (DOE). For the category of "Energy Performance", efforts will be directed to environmentally benign, low-cost energy sources, including recovery and gasification of process residues

Forestry biomass fuels contain typically 40-65 % moisture by weight. Current IGCC technology is suited to the use of low moisture content fuels. A moisture content of 11% is used in high pressure fluidised bed gasification technology (PDU, Craig and Mann, 1997). This is achieved in a co-current rotary drum drier utilising process heat. The traditional method of obtaining a low moisture content fuel is to use waste heat from the IGCC process to dry the biomass prior to gasification. The difficulty with this is that the forestry industry needs the waste heat for other processes, eg. timber and paper drying.

In designing an IGCC system for a pulping company, the following features are desirable:

- Ability to utilise fossil fuels to meet the shortfall in the bio-energy supply
- High electrical efficiency
- Efficient utilisation of the waste heat
- Ability to use biomass fuels with high moisture contents
- Flexibility to cope with variations in fuel supply
- Competitive capital cost
- Low maintenance costs
- Low labour input

6.2 Method

6.2.1 High moisture content fuels

Gas analysis was conducted by Tony Clemens (CRL) and Steve Goldthorpe (WWC). The method and results of this analysis is contained in “Analysis of gas from a pressurised air blown downdraft biomass gasifier” (Clemens and Gong, 1998) and “Massey Gasifier Tests” (Goldthorpe, 1998). Experimental assistance to help run the gasifier was provided by Badenoch Sime.

A bed length of 10 cm was used for all of the experiments. Experiments were done at both high pressure (600 kPa) and low pressure (100 - 200 kPa). A volume flow rate of approximately 25 l.s^{-1} was used.

The fuels used for this study were:-

- Wood-chip, 4% m.c by wt.
- Wood-chip, 18% m.c by wt.
- Wood-chip, 45% m.c by wt.
- Bark, 9% m.c by wt.
- Bark, 43% m.c by wt.

The wood-chip and bark used in this study are shown in Figures 6.1 and 6.2 respectively. Wood-chip (45% m.c.) was gasified by starting the gasifier on wood-chip (18% m.c.) followed by wood-chip (45% m.c.).

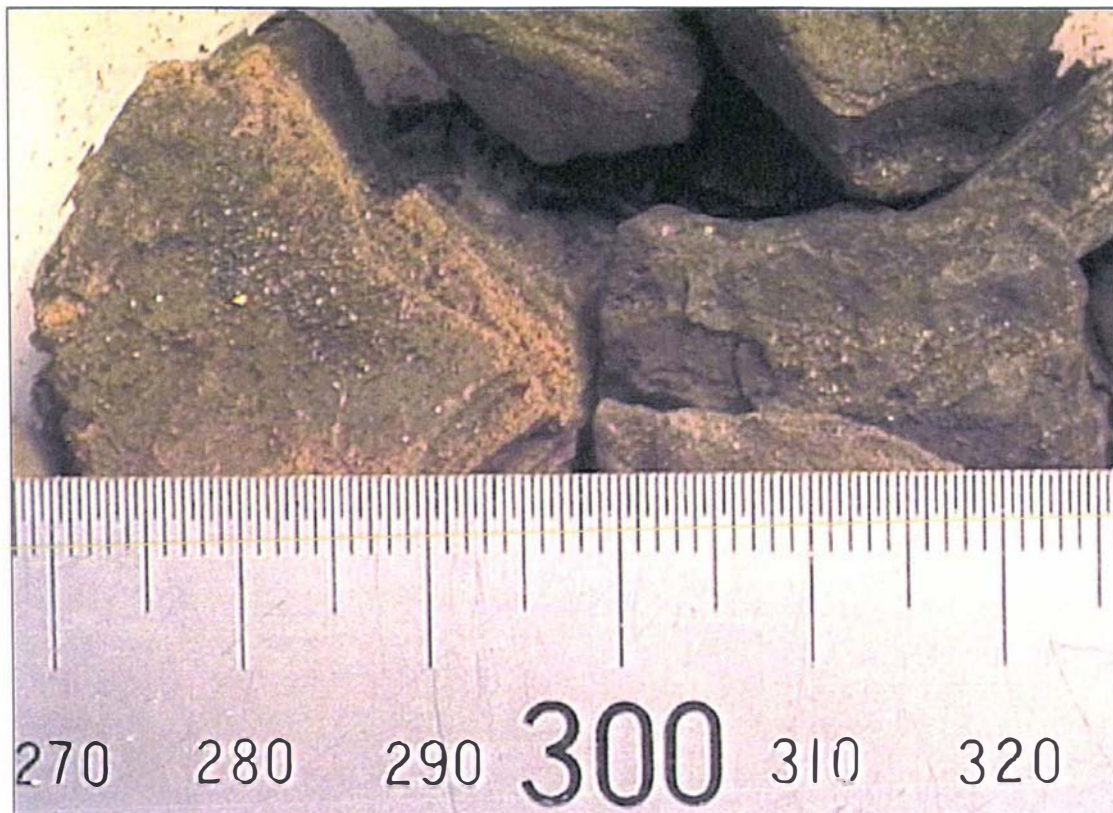
Figure 6.1: Wood-chip



An attempt was made to start the gasifier on wood-chip (45% m.c.), followed by bark (43% m.c.). The gasifier did not start properly, which caused partial

melting of the exit valve, exit pipe and burner, preventing further experimental work.

Figure 6.2: Bark



6.2.2 Bark Mulch

The gasifier was loaded with half wood-chip (4% m.c.) on the bottom, followed by bark mulch on the top. The gasifier was started and monitored in the normal way. The bark mulch did not start to gasify until mid way through the gasifier run. The gasifier pressure and volume flow rate were 910 kPa. and 23 l.s^{-1} respectively.

Figure 6.3: Bark Mulch 6% m.c.



Figure 6.4 Left; bark, Top right; bark mulch, Bottom right; wood-chip



6.3 Results and Discussion

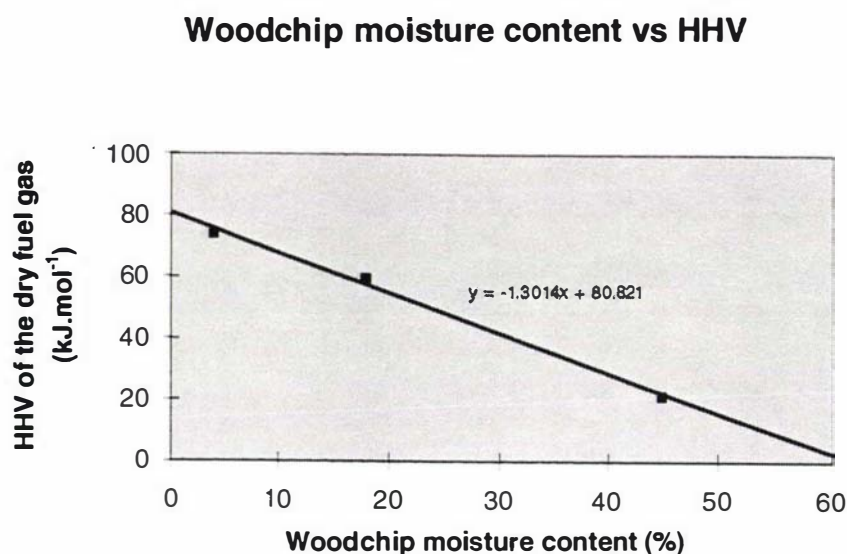
6.3.1 Effect of moisture on the fuel-gas

Mobile gas chromatography (g.c.) equipment was brought from Coal Research Limited to the gasifier for the gas analysis. This was the first time that both the gas chromatograph and gasifier had been coupled together. Initially the sampling system which interfaced the two sets of equipment had some air leaks. The most significant of these leaks were sealed, and the tiny amount of remaining air was subtracted using software. The g.c. thermally equilibrated quickly and was very stable. The data collection time from the gasifier and the g.c. were matched so that the data could be compared.

In Figure 6.5 the effect of wood-chip moisture content on the fuel-gas energy content is shown. Data for Figure 6.5 had the following characteristics:

- Wood-chip m.c. 4%, 18% and 45%
- Volume flow rate: $25 \pm 3 \text{ l.s}^{-1}$
- Pressure: $660 \pm 20 \text{ kPa}$
- Bed length 10 cm
- Minimum exit temperature 1300 K

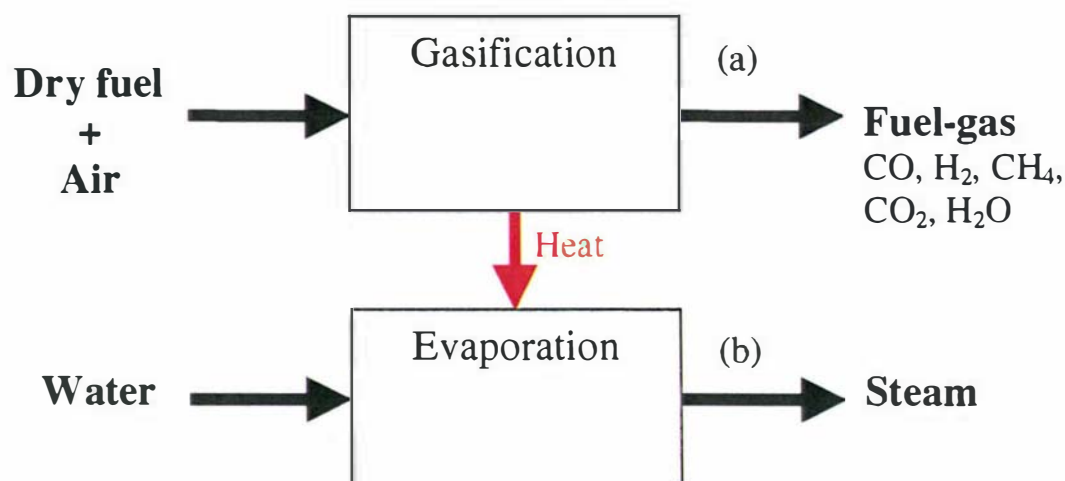
Figure 6.5



The main woody fuel produced at pulp mills is bark mulch. Bark mulch is a mixture of bark and white wood, which is removed from the outside of the log during debarking. Sometimes this is mixed with saw dust from local saw mills. At the local pulp mill (Kinleith) about 10-15% of the bio-fuel used in the combustor is saw dust, the remainder is bark mulch. Forestry biomass fuels contain typically 40-65 % moisture by weight.

In gasification the chemical energy of the wood is transformed to chemical energy of the fuel-gas and sensible energy. If the fuel is wet, some energy is used heating the water to form high temperature steam (Figure 6.6). Water has the effect of decreasing the fuel-gas chemical energy and increasing the sensible energy. This is in agreement with the experimental observations (Figure 6.5).

Figure 6.6



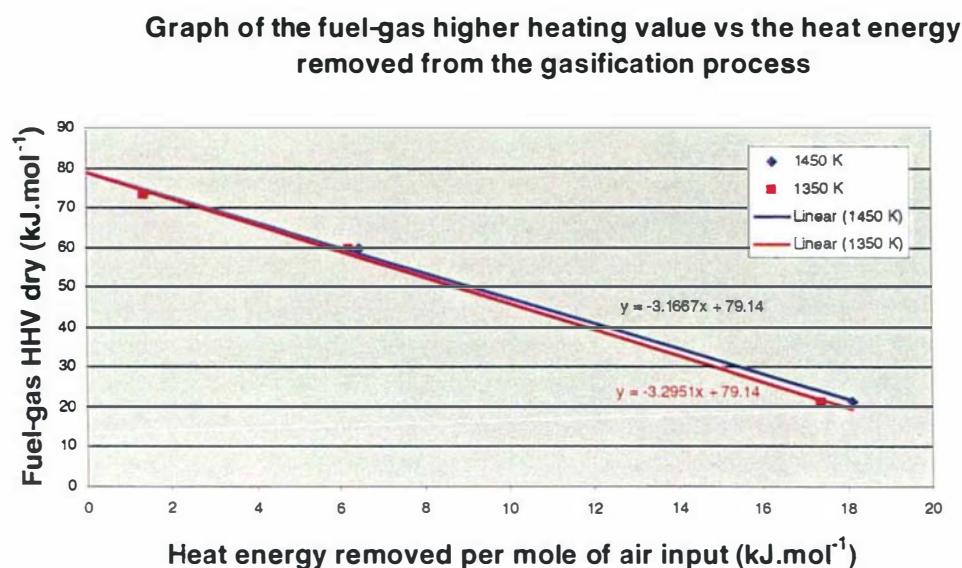
Treating the moisture in the wood fuel as an energy loss, it is possible to calculate the effect of heat energy addition or subtraction from gasifier. To do this the following steps are taken. For conceptual simplicity the wood moisture is treated as being separated through a heat exchanger from the gasification process of the dry wood (Figure 6.6).

From the gas chromatography data, the typical composition of wood ($\text{CH}_{1.4}\text{O}_{0.6}$) and moisture content of the wood, the following may be calculated:

1. The mole fraction of H_2O boiled off from the wood moisture (stream (b))
2. The mole fraction H_2O from combustion (stream (a))
3. The molar expansion of the gas stream (a)
4. The amount of energy required to boil off the water in stream (b) per mole of air entering the gasifier (at exit temperatures 1350 K and 1450K)
5. The higher heating value of fuel-gas (stream (a))

The exit temperature for this data is not known but was at least 1350 K. Therefore analysis has been done at 1350 K and 1450 K. A graph of the fuel-gas dry higher heating value vs the heat removed per mole of air input is shown in Figure 6.7.

Figure 6.7

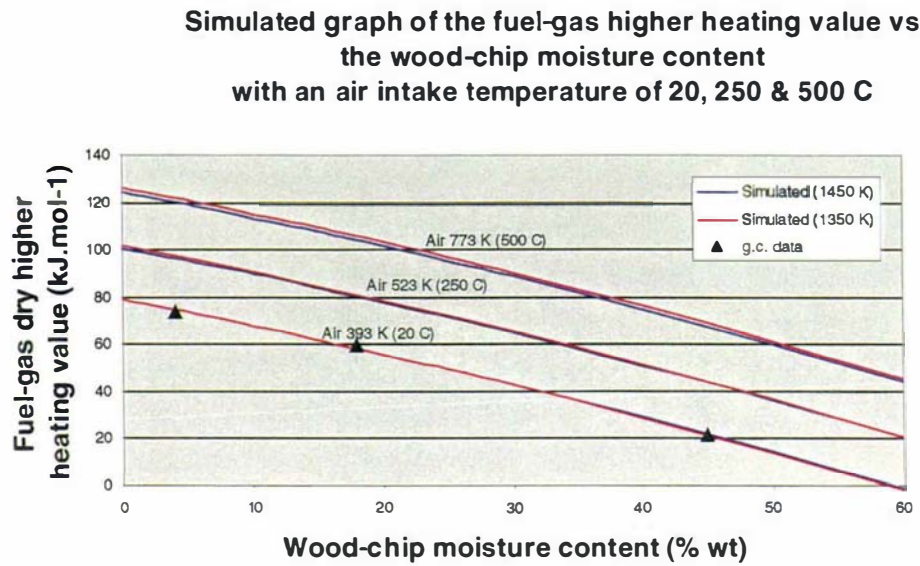


From Figure 6.7 it can be seen that the exit temperature had little effect on the heat energy removed per mole of air input. It is possible to add heat energy into the gasification system by pre-heating the intake air. The intake air can be pre-heated using a combination of the following methods:

1. Adiabatic compression of the intake air by the gas turbine
2. Transfer of energy to the intake air from the exit gas stream.

The intake air temperature could potentially be increased to 1300 K if it was desired. Predictions can be made about how pre-heating the intake air would effect the fuel-gas heating value. The process is simple since both energy addition and removal can be quantified in terms of heat per mole of intake air. In Figure 6.8 the effect of moisture content on the higher heating of the fuel-air is simulated for an intake air temperature of 250 °C (523 K) and 500 °C (773 K). Real gas chromatography data and simulated data at 20 °C (373 K) is shown for comparison.

Figure 6.8



6.3.2 Dry fuels

Traditional high pressure IGCC technology utilises a fuel with a moisture content of 11% (PDU, Craig and Mann, 1997). These systems use rotary drum dryers to dry the fuel. The energy for drying the fuel is supplied as heat from the IGCC plant. Using simulated data obtained from Figure 6.8 at an intake air temperature of 773 K (500 C) a dry fuel-gas energy content of 112.5 kJ.mol⁻¹ is possible. Gas turbine combustors can be designed to run on a fuel-gas HHV of 100 kJ.mol⁻¹ without much difficulty. An IGCC system could be designed to run using heat to pre-dry the fuel. A disadvantage of using heat to pre-dry the fuel is that heat is used for other processes in the forestry industry.

6.3.3 Wet fuels

The ability to use wet fuels is an advantage when designing an energy system for pulp mills. The traditional energy systems used in these mills are combustion systems. These combustors burn wet fuel (40–65 % wt), often in combination with fossil fuels like natural gas, fuel oil and coal.

At high moisture contents (Figure 6.8, air intake 773 K) the fuel-gas higher heating value is too low to run a gas turbine (HHV dry $\ll 100 \text{ kJ.mol}^{-1}$). One option is to further increase the intake temperature, thus increasing the fuel-gas HHV. A more practical method may be to utilise the fact that in many pulp mills, fossil energy is often required. The fossil fuels could be used to boost the HHV of the gas stream, thus enabling the gas turbine to function.

Another option is to develop Integrated Drying and Gasification Combined Cycle (IDGCC) technology for biomass fuels. IDGCC technology is currently being developed for gasification of wet coal fuels (Anderson *et. al.*, 1998). In IDGCC the gasifier fuel-gas exit stream is mixed with wet coal. The water in the coal evaporates cooling the fuel-gas stream. This method has the advantage the heat of vaporisation of the coal moisture is retained in the energy system. Proponents of the technology claim IDGCC is a cheap alternative to using heat exchangers and conventional coal dryers. It might be possible to develop IDGCC technology for biomass fuels.

6.3.4 Boosting with natural gas

A popular fossil fuel choice in the pulp and paper industry is natural gas (Table 6.1). The following scheme could be used to co-fire biomass fuel-gas and natural gas into a gas turbine.

- Run the biomass fuel-gas into the gas turbine combustor at the same pressure as the air.
- The natural gas is supplied at a higher pressure than the gas turbine combustor.
- Regulation is primarily controlled by varying the supply of natural gas.
- Combustor design is very simple. Use a standard natural gas burner for initial combustion and regulation. The natural gas burner will act as a vastly oversized pilot burner for the biomass fuel-gas.
- A gas turbine which uses moisturised combustion technology would be a good first choice (see IGAC section 1.3.6). The moisture (steam) for this type of gas turbines is normally supplied primarily from the recuperator. When using wet biomass fuels, moisture will also be supplied from the gasifier. Excess steam from the recuperator can be used for co-generation applications.

6.3.5 Boosting with coal

Coal is another popular fossil fuel choice in the pulp industry. Coal fuel-gas could also be used to compensate for the low HHV of fuel-gas from wet biomass. There are a number of possible configurations:

1. Co-gasifier with coal and wet biomass in the same gasifier. This method may prove to be very demanding, as the two fuels are very different in their properties.
2. Gasify the coal and wet biomass in separate gasifiers. This is an attractive option but the capital cost is increased. The coal and biomass fuel-gases could be mixed prior to filtration. Alternatively the higher energy coal fuel-gas could be fired separately into the gas turbine combustor to ensure good initial combustion.

6.3.6 Black-liquor

Sodium hydroxide along with dihydrogen sulfide (H_2S) is used in the chemical pulping of wood-chips. Lignin is dissolved to form sodium salts (black-liquor), leaving the cellulose fibres unreacted. The black-liquor is then combusted in a furnace to recover the process chemicals and provide energy for the pulping plant.

If designing an energy system for a pulp mill, how best to utilise the black-liquor might also be considered. Gasification technology is being developed for black-liquor fuels. Sodium is recovered from the black liquor at the gasifier (as oxides of sodium), whereas sulfur is recovered as dihydrogen sulfide from the fuel-gas stream using hot gas desulphurisation technology.

It is claimed by proponents of the technology that black liquor gasification technology used in conjunction with gas turbine co-generation systems has the following advantages over traditional recovery furnaces:

- Higher overall energy efficiency
- Lower volume of gas requiring treatment
- Lower emissions (both gaseous and particulate)
- Higher inherent safety; no explosion hazard posed by molten smelt, which is present in recovery furnaces
- Higher adaptability to handling variations in liquor capacity by using multiple gasifiers or by other process modifications

Proponents also assert that black liquor gasification will become economically competitive with traditional recovery furnaces within the next few decades.

Black liquor is the most important bio-energy used in the pulp mills. It typically provides three times as much energy as woody bio-residues. Bark and wood residues make up only 15% of the total carbon based energy used in U.S. mills (Table 7.1). If black liquor is not included, woody bioenergy is still only 28% of the carbon energy contribution.

Ultimately systems may be designed using gas turbine technology, which take fuel from all of the carbon based energy sources. Fuel-gas from woody residues will make only a minor contribution to the total fuel-gas flow. The low chemical energy content of fuel-gas from wet woody residues will not be of any consequence when boosted by the bulk flow of fuel-gas from other energy sources. Therefore, in designing a gasification system for pulp mills, the use of wet fuels should be a priority.

6.3.7 Bark mulch

Bark mulch is produced in pulp mills when the logs are debarked. Bark mulch is a mix of bark and wood residues. It is a fuel with a high fines content. The traditional atmospheric pressure downdraft gasifier does not normally use this fuel. To quote from a gasification expert (Kurkela et. al., 1993)"The fixed bed downdraft gasifier is suitable for small scale engine applications with dense, high quality feed-stocks such as pellets, briquettes, hog fuel, or high-quality wood chips". They go on to say that feed-stocks for a downdraft gasifier should have "very low fines content". A significant reason why bark mulch is not normally used is that the gasifier is more likely to block when using fuels containing a high fines content.

The gasifier was loaded with half wood-chip (4% m.c.) on the bottom, followed by bark on the top. The bark mulch did not start to gasify until mid way through the gasifier run. The hopper was checked at the end of the run to confirm that the gasifier had been using bark-mulch fuel. Details of the run are shown in Table 6.2. The gasifier functioned well on bark mulch with no observed problems.

The low bed pressure drop (Table 6.2) indicates that blockage was not an issue. When low exit temperatures were used in Chapter 4, even top quality fuels such as wood-chip caused blockage. Running the gasifier with high exit temperatures appears to be a key component in increasing fuel flexibility.

Table 6.2

Parameter	Value
Pressure	910 kPa
Volume flow rate	23 l.s ⁻¹
Molar flow rate	8.7 mol.s ⁻¹
Bed pressure drop	0.3 kPa
Bed length	12 cm
Moisture content	6 % wt
Expected thermal output	37 MW _t .m ⁻²

In addition it is possible that waste timber residues such as saw dust and shavings might be blended in with the bark mulch. This increased fuel flexibility may allow the technology to be suitable for pulp mills. More detailed investigation is required to properly quantify the suitability of bark mulch as a fuel, but preliminary indications are very encouraging and well surpass initial expectations of the technology.

6.3.8 Bark

The bark used in this study was obtained from the local garden centre. Being a pure source of chip size bark, it has a higher heating value than bark mulch. It is unlikely that a pure bark fuel will be used in a commercial energy plant.

Bark was used in the gasifier at both high and low pressure. The fuel gas energy content (55-62 kJ.mol⁻¹) was lower than expected from comparison to wood-chip of that moisture content (68 kJ.mol⁻¹, Figure 6.5). Since bark normally has a slightly higher energy content than wood, this is difficult to

account for. Perhaps it might be attributed to the bark being old and somewhat composted.

6.3.9 Coal

Coal is a dominant fuel for World electricity production. As a fuel, coal is far more significant than biomass. Much of the current IGCC technology that is utilised for biomass fuels was originally developed for coal fuels. Coal IGCC technology such as in the Pinion Pine plant is already at the commercial demonstration stage.

Gasifier capital costs currently account for a quarter of the cost of producing electricity from Coal IGCC (FETC, 1997). Reducing gasifier capital cost is considered to be a key component to advancing the commercial success of Coal IGCC (FETC, 1997). For this reason, the U.S. government is currently funding 5 different gasifier options. These gasifier options include fluidised bed, entrained flow and transport (cyclonic fluidised bed). Furthermore there are many other worldwide initiatives.

It is possible to design gasification technology that is suitable for either woody biomass or coal. For example, fluidised bed gasifiers can be used for both coal and woody biomass. Coal is a less reactive fuel than woody biomass. To get comparable reaction rates from coal, higher temperatures are required. For example, by running the gasifier with an exit temperature 400 K higher than the traditional biomass downdraft gasifier (1025 K), much higher surface kinetic reaction rates will be possible (Figure 4.9).

The use of coal has been briefly trialed. The coal used, was supplied in 10 kg bags produced by Solid Energy. Data for the run is shown in Table 6.3. Coal gasified well, with a good stable burner flame. Further investigation will be

required to make an informed recommendation on the suitability of the HTP downdraft for gasifying coal fuels. Gasification of coal is an interesting research opportunity, which should be explored.

Table 6.3

Parameter	Value
Pressure	400 kPa
Volume flow rate	32 l.s ⁻¹
Molar flow rate	5.1 mol.s ⁻¹
Bed pressure drop	0.5 kPa
Bed length	12 cm

6.3.10 Start up problems with wet fuels

During run number 1806-6, the gasifier contained wet wood-chip (m.c. 45%) from the previous run. Wet bark (m.c. 43%) was loaded on top. The gasifier did not start well with the exit temperature very low. The gasifier was then stopped and another attempt was made (run 1806-7). This time the gasifier did start to heat up. After about 500 seconds metal started to hit the outlet burner nozzle. The volume flow rate increased from 18 l.s⁻¹ up to 35 l.s⁻¹. After 580 seconds, a hole developed in the side of the exit pipe and valve, the volume flow rate increased to 60 l.s⁻¹. Within seconds of the hole developing, the gasifier was shut down.

Inspection of the gasifier found:

- The burner had a hole and was coated inside with a molten stainless spray (Figure 6.8)
- The high temperature exit valve had no internal components remaining (Figure 6.9)
- A hole had melted in the side of the exit pipe and valve (Figure 6.10)
- The gas sampling probe (stainless steel) had the end melted off
- The gasifier bed had unburnt bark right down to the level of the grate
- The grate was in perfect working order
- The grate vertical support shaft (25 mm high temperature stainless) was melted 10 cm below the grate on one side only.
- The stainless thermocouple sheath was fine
- The cyclone blades were fine, but the central exiting pathway was melted in two places.

Figure 6.8 Burner nozzle

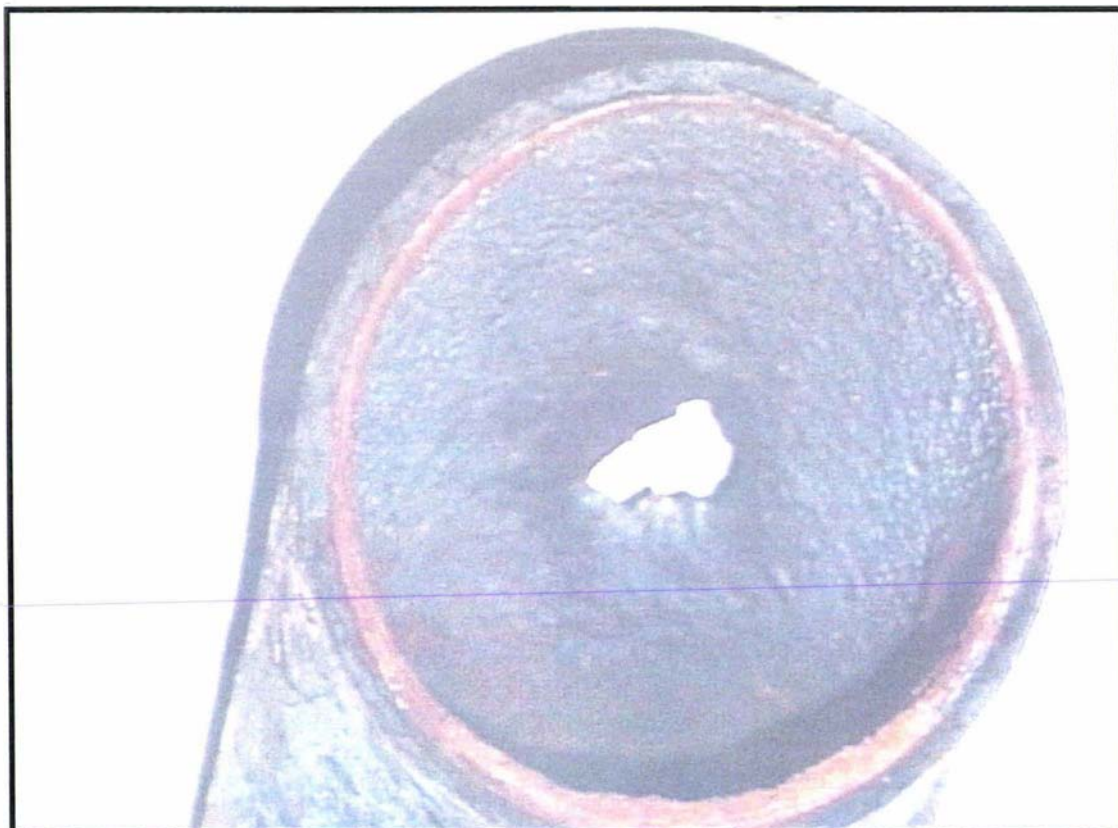


Figure 6.9 High temperature valve

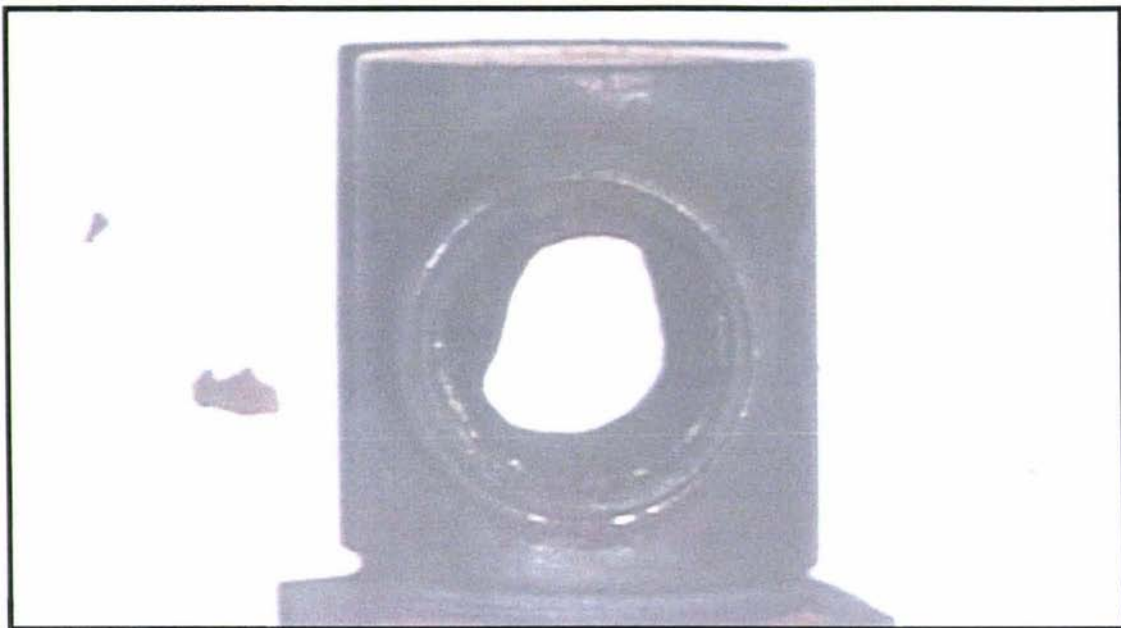


Figure 6.10 High temperature exit valve and pipe



From examination of the evidence, it would appear that combustion of fuel gas and air occurred in the gasifier, raising the temperature above the melting temperature of high temperature stainless steel. The melting occurred only in localised regions where gases exiting from the bed had mixed. Air managed to get through the bed uncombusted because part of the bed had gone out. The gasifier had failed to start properly on wet fuels.

6.3.11 Prevention of start up problems

Fuels of high moisture content (56 %) have been used, when the gasifier is first started on dry fuel. Downdraft gasifiers normally combust with high intensity at the position of inlet nozzles. This provides heat for the gasification process. In a normally operating gasifier, the fuel is pre-dried by the heat from combustion before arriving at the nozzles. During start up, insufficient heat may be available to pre-dry the fuel. The result is that wet fuel may sit in front of the nozzles entrained in a cold gas stream (air cooling).

In the real IGCC system the air entering the gasifier is at 800 K (500 °C). Not only will this cause rapid drying, fast pyrolysis and char production will also occur. Fast biomass pyrolysisers operate at 800 K. Therefore this melting phenomenon can not occur during continuous operation in an IGCC system.

It would be advisable when starting with wet fuels that natural gas is combusted at the air nozzles around the circumference of the bed. This would ensure proper gasifier start up. After the gasifier has warmed up, the natural gas could be shut off. In addition thermocouples could be placed around the circumference of the gasifier bed, below the air nozzles. A low temperature will indicate that the bed has not ignited correctly

6.4 Conclusion

The main market for woody bio-energy technology is the existing pulp industry. In this industry, heat is valuable. Using heat to dry the fuels is undesirable. Gasifying wet fuels results in a low fuel-gas chemical energy content, which is too low to power a gas turbine (even with air pre-heating). The fuel-gas energy content may be boosted with another fuel source. Other possible fuel sources used at pulp mills include, natural gas, coal and black-liquor. If wet fuels are used, additional starting technology must be installed to avert start-up problems.

Another possibility is to utilise IDGCC technology. If IDGCC technology can be applied to biomass fuels, then the valuable heat used for drying the fuel will not be lost from the energy system. IDGCC is well suited to co-generation applications.

Successful gasification of bark mulch was one of the highlights of the project. Bark mulch will be used in future experimental work. The possibility of experimental coal gasification was also demonstrated. Further research using coal fuels is recommended.

7.0 Overall conclusion

The downdraft gasifier first entered into widespread use during World War One. At the time the main use was as mobile installations on motor vehicles. Since then there have been many downdraft gasifier variations.

Over the last century downdraft gasifiers have been gradually improving. There has been significant advancement in high temperature construction materials, automation of the fuel feed, and automated char removal systems. Downdraft gasifiers are a proven technology, known for their simplicity and better gas quality.

In recent years the usefulness of the traditional downdraft gasifier has been displaced by advanced fluidised bed technologies. The chief advantages of fluidised bed gasifiers are that they are suited to medium to large scale applications and have good fuel flexibility. For the downdraft to be a competitive technology, larger throughputs and better fuel flexibility are essential.

Research was initiated by considering the effect of pressure and gas flow on the gasifier bed. This was done by mathematically treating the gasifier bed as a typical bed of solids. From analysis of the mathematical model it was hypothesised that pressure could be used to increase the gasifier throughput.

Non-equilibrium experimental work confirmed that pressure could be advantageously utilised to aid throughput. It also showed another effect, that the particle distribution gave less resistance to gas flow at high exit temperatures.

In the traditional downdraft gasifier, there is an excess of bed material. As the gas stream passes through the bed gasification reactions continue until

insufficient heat energy remains. The excess bed material (sometimes called the dead zone) is prone to blocking with fine char particles. Traditionally this problem is solved using moving grates that physically remove the char from the dead zone.

It was found that by running the gasifier with a short bed length increased the exit temperature of the fuel-gas. In doing this the dead zone is removed, hence automated char removal systems are not required. Another advantage is that the bed is free flowing with only low pressure drops. Hence, running the gasifier with a high exit temperature is another parameter that can be used to increase throughput.

High temperature and pressure aid tar cracking. Therefore the HTP downdraft is expected to have better tar cracking ability than the traditional downdraft gasifier. A disadvantage of high exit temperatures is that the fuel-gas chemical energy content is reduced. This can be compensated for by increasing the intake air temperature.

The current market for woody bioenergy technology is primarily in the pulp mills. In this industry process heat is used on a large scale. Therefore the current market is almost entirely co-generation. The woody fuel is normally combusted wet.

Gasification with wet fuels was investigated. It was found that high moisture content decreased the fuel-gas energy content. Within the pulp industry woody biomass makes only minor carbon based energy contribution (15%). It is possible to boost the fuel-gas energy content using other fuel-gases. Therefore the low fuel-gas chemical energy content is of little consequence. Further experimentation with wet fuels is recommended. The HTP downdraft gasifier will require the development of start-up technology to avoid component failure when using high moisture content fuels.

The HTP gasifier has been only run for short periods of time. Pressures up to 900 kPa using air supplied from an isothermal compressor have been investigated. Continued research with long gasifier runs, pressures up to 2500kPa and intake air temperatures of at least 800 K is advised.

Experimental results have confirmed the expectation that the throughput of HTP downdraft gasifier will far exceed that of the traditional downdraft gasifier. The improved fuel flexibility allows gasification of difficult fuels such as bark mulch. Coal gasification is also a possibility.

Capital costs for the HTP downdraft gasifiers are expected to be very low, less than 10% of the competitive fluidised bed technology. Low HTP gasifier cost (\$3000 so far) has had the advantage that the research investment to date has been very low. The cost of continued research will remain very modest.

In conclusion, the HTP downdraft gasifier overcomes the tradition downdraft problems of low throughput and poor fuel flexibility. The HTP downdraft gasifier is a low cost alternative to the current fluidised bed gasifiers.

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9.0 Appendix: Gas Chromatography data

These were collected by Coal Research Limited during the fuel flexibility experiments (chapter 6). They should be interpreted in conjunction with the logged gasifier parameters, which may be obtained from the author, or alternatively from Dr Geoffrey Barnes.

1706-1 cold		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 17, 199	1	0.2	0.49	0	0.9	18.69	76.98	1.73
Jun 17, 199	2	0.2	0.5	0	0.81	18.73	76.61	1.62
Jun 17, 199	3	0.22	0.56	0	0.79	18.81	76.95	1.65
Jun 17, 199	4	0.23	0.59	0	0.71	18.62	75.81	1.5
Jun 17, 199	5	0.22	0.6	0	0.62	19.14	77.06	1.41

1706-3		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 17, 198	1	0	0.04	0	0	20.92	79.1	0
Jun 17, 198	2	0.57	4.79	0	7.82	0.66	67.89	15.57
Jun 17, 198	3	0.65	9.7	0	7.91	0.17	65.31	15.3
Jun 17, 198	4	0.47	10.63	0	8.2	0.26	62.39	15.86
Jun 17, 198	5	0.97	9.29	0	2.14	14.26	67.77	5.13
Jun 17, 198	6	0.01	3.13	0	0.01	20.04	75.9	0
Jun 17, 198	7	0	1.49	0	0	20.41	77.15	0

1806-3		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 18, 199	1	0	0.05	0	0	20.7	78.21	0
Jun 18, 199	2	0.09	0.91	0	4.13	7.29	76.53	8.38
Jun 18, 199	3	0.08	9.07	0	5.9	0.21	68.33	13.61
Jun 18, 199	4	0.03	10.88	0	6.03	0.19	67.22	12.29
Jun 18, 199	5	0.04	11.5	0	6.92	0.23	65.52	12.36
Jun 18, 199	6	0.14	11.64	0	7.65	0.24	64.35	13.27
Jun 18, 199	7	0.21	11.56	0	8.27	0.25	62.8	13.39
Jun 18, 199	8	0.75	10.42	0	1.86	14.96	67.01	2.71

1706-4		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 17, 199	1	0	0.3	0.01	0.01	20.64	78.2	0
Jun 17, 199	2	0	0.13	0	0.05	20.75	78.56	0
Jun 17, 199	3	0.41	1.43	0	1.51	16.49	74.5	3.61
Jun 17, 199	4	0.41	1.77	0.01	1.3	16.98	74.45	3.17
Jun 17, 199	5	0.26	1.64	0	1.06	17.73	75.05	2.37
Jun 17, 199	6	0.25	1.55	0	1.01	17.88	75.23	2.22
Jun 17, 199	7	0.19	1.49	0	1	17.9	75.21	1.97
Jun 17, 199	8	0.2	1.6	0	1.14	17.95	75.91	2.15
Jun 17, 199	9	1.59	4.03	0	7.76	1.93	65.97	16.78
Jun 17, 199	10	1.5	7.67	0	7.54	1.37	62.23	17.09
Jun 17, 199	11	1.27	9.21	0	7.64	1.26	61.62	16.56
Jun 17, 199	12	1.18	9.94	0	7.69	1.36	61.57	15.59
Jun 17, 199	13	1.5	10.48	0	7.92	1.23	60.08	16.17
Jun 17, 199	14	1.31	11.72	0	6.87	0.21	61.91	15.92
Jun 17, 199	15	1.57	11.96	0.01	7.24	0.49	59.94	16.47
Jun 17, 199	16	1.8	11.51	0.01	7.21	0.7	58.42	17.27
Jun 17, 199	17	1.91	11.22	0.01	6.79	0.86	59.16	17.05
Jun 17, 199	18	1.9	10.93	0.01	7.11	1.51	58.28	16.61
Jun 17, 199	19	1.2	5.56	0.01	2.04	17.31	67.95	2.94
Jun 17, 199	20	0.28	5.54	0	0.26	19.08	72.36	0.56
Jun 17, 199	21	0	2.37	0.01	0.01	20.08	75.59	0
Jun 17, 199	22	0	1.25	0.01	0.01	20.27	76.29	0
Jun 17, 199	23	0	0.79	0.01	0.01	20.43	76.98	0
Jun 17, 199	24	0	0.7	0	0.01	20.37	76.71	0
Jun 17, 199	25	0	0.39	0.01	0	20.4	76.77	0
Jun 17, 199	26	0	0.26	0.01	0	20.44	77.14	0

1706-5.dif	999	Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
Jun 17, 199	1	0	0.036	0	0.005	20.967	79.003	0
Jun 17, 199	2	0	0.035	0	0.001	20.843	78.569	0
Jun 17, 199	3	1.284	5.23	0	7.424	0.911	67.937	14.526
Jun 17, 199	4	1.144	10.739	0	7.02	0.121	64.767	14.668
Jun 17, 199	5	1.01	11.617	0	7.255	0.117	63.012	14.97
Jun 17, 199	6	0.957	12.159	0	7.97	0.216	61.521	15.022
Jun 17, 199	7	0.789	12.012	0	7.914	0.105	61.593	15.869
Jun 17, 199	8	0.44	12.261	0	6.379	0.12	64.66	13.483
Jun 17, 199	9	1.591	12.409	0.004	6.292	5.377	62.119	10.848
Jun 17, 199	10	0.018	4.329	0.005	0.016	19.601	74.217	0.06

1706-6

		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 17, 198	1	0	0.04	0	0	20.91	78.83	0
Jun 17, 198	2	0	0.05	0	0.4	19.5	77.95	0.68
Jun 17, 198	3	0.13	8.49	0	4.83	0.35	70.71	13.35
Jun 17, 198	4	0.13	10.67	0	5.71	0.51	66.39	14.32
Jun 17, 198	5	0.56	8.64	0	1.34	16.02	69.39	2.77

1806-0		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 18, 198	1	0	0.04	0	0	21.22	80.09	0
Jun 18, 198	2	0	0.04	0	0	20.83	78.83	0
Jun 18, 198	3	0	0.04	0	0	20.95	79.29	0
Jun 18, 198	4	0.01	0.61	0	0.4	19.18	78.94	0.93
Jun 18, 198	5	0	0.55	0	0.01	20.86	78.67	0
Jun 18, 198	6	0	0.32	0	0	20.81	78.69	0
Jun 18, 198	7	0	0.2	0	0	20.64	78.1	0
Jun 18, 198	8	0	0.14	0	0	20.9	79.06	0
Jun 18, 198	9	0	0.1	0	0	20.81	78.74	0
Jun 18, 198	10	0.02	0.15	0	0.09	20.54	78.89	0
Jun 18, 198	11	0	0.17	0	0.02	20.73	78.34	0
Jun 18, 198	12	0	0.06	0	0.01	20.81	78.39	0
Jun 18, 198	13	0	0.04	0	0	20.86	78.54	0
Jun 18, 198	14	0	0.08	0	0.15	20.49	78.66	0.08
Jun 18, 198	15	0.01	0.08	0	0.15	20.26	78.89	0.2
Jun 18, 198	16	0.01	0.1	0	0.14	20.26	78.56	0.02
Jun 18, 198	17	0.01	0.12	0	0.14	20.28	78.48	0.13
Jun 18, 198	18	0.01	0.15	0	0.13	20.34	78.61	0.22
Jun 18, 198	19	0.02	0.19	0	0.15	20.27	78.39	0.07

1806-1		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 18, 199	1	0	0.04	0	0	20.95	79.34	0
Jun 18, 199	2	0	0.04	0	0	20.9	79.26	0
Jun 18, 199	3	0	0.04	0	0	20.77	78.83	0

		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 18, 199	1	0	0.04	0	0	20.79	78.68	0
Jun 18, 199	2	0	0.04	0	0	20.75	78.82	0
Jun 18, 199	3	0.71	6.03	0	4.65	0.39	71.93	14.26
Jun 18, 199	4	0.38	9.6	0	4.79	0.22	70.01	14.01
Jun 18, 199	5	0.29	10.56	0	4.99	0.21	68.1	13.79
Jun 18, 199	6	0.32	11.04	0	5.52	0.24	67.27	14.06
Jun 18, 199	7	0.37	11.03	0	6.23	0.24	65.81	14.81
Jun 18, 199	8	0.4	11.44	0	5.94	0.11	67.33	13.17
Jun 18, 199	9	0.48	12.11	0	6.54	0.12	65.5	13.77
Jun 18, 199	10	0.33	12.33	0	6.37	0.22	65.57	13.63
Jun 18, 199	11	0.31	11.63	0	6.54	0.2	63.78	15.79
Jun 18, 199	12	0.36	11.23	0	6.9	0.1	64.53	15.26
Jun 18, 199	13	0.53	11.85	0	6.61	0.29	65.24	13.55
Jun 18, 199	14	0.52	11.54	0	7.24	0.19	62.92	15.3
Jun 18, 199	15	0.51	11.69	0	7.4	0.26	64.15	13.99
Jun 18, 199	16	0.63	12.08	0	7.06	0.2	63.4	14.51
Jun 18, 199	17	0.45	11.34	0	3.28	10.05	66.59	7.67
Jun 18, 199	18	0.02	4.25	0	0.12	19.57	74.9	0.08

1806-2		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 18, 195	1	0	0.04	0	0	20.86	78.86	0
Jun 18, 195	2	0.12	2.07	0	5.02	2.14	75.51	13
Jun 18, 195	3	0.16	8.57	0	5.56	0.13	69.44	13.95
Jun 18, 195	4	0.18	11.02	0	5.14	0.12	69.05	12.43
Jun 18, 195	5	0.08	11.69	0	5.9	0.12	66.9	12.78
Jun 18, 195	6	0.05	11.6	0	5.45	0.12	66.52	13.36
Jun 18, 195	7	0.03	12.07	0	5.66	0.13	67.47	11.47
Jun 18, 195	8	0.02	12.13	0	6.11	0.14	66.62	11.92
Jun 18, 195	9	0.92	11.8	0	12.72	0.2	57.37	14.28
Jun 18, 195	10	0.03	5.09	0	0.03	19.51	74.04	0

1806-4		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 18, 19!	1	0	0.04	0	0	20.52	77.43	0
Jun 18, 19!	2	0.01	0.05	0	0.34	19.77	77.77	0.09
Jun 18, 19!	3	0.87	11.31	0	7.36	0.29	65.81	10.17
Jun 18, 19!	4	0.73	12.99	0	7.09	0.18	64.48	10.22
Jun 18, 19!	5	0.97	13.52	0	7.63	0.21	63.52	10.68
Jun 18, 19!	6	0.08	14.86	0	3.07	0.19	73.95	3.92
Jun 18, 19!	7	0.12	15.99	0	3.18	0.22	72.44	3.55
Jun 18, 19!	8	0.01	5.04	0	0.01	19.11	72.62	0
Jun 18, 19!	9	0	1.89	0.01	0	19.8	75.03	0

1806-5		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 18, 19	1	0	0.04	0	0	20.59	77.64	0
Jun 18, 19	2	0.19	1.39	0	1.76	7.78	85.55	1.53
Jun 18, 19	3	0	14.4	0	0.02	0.83	82.4	0
Jun 18, 19	4	0	14.53	0	0	4.4	78.5	0
Jun 18, 19	5	0	13.38	0	0	6.25	78.03	0
Jun 18, 19	6	0	13.01	0.01	0	5.36	78.77	0
Jun 18, 19	7	0	16.08	0	0	2.12	78.69	0
Jun 18, 19	8	0.07	17.07	0.01	0.82	2.32	76.3	0.22
Jun 18, 19	9	0	5.73	0.01	0	19.13	72.61	0
Jun 18, 19	10	0	2.35	0	0	19.9	75.26	0

1806-6		Methane	Carbon Dic	Ethane	Hydrogen	Oxygen	Nitrogen	Carbon Mo
		%	%	%	%	%	%	%
Jun 18, 195	1	0	0.04	0	0	20.74	78.27	0
Jun 18, 195	2	0	0.1	0	0.2	18.75	79.4	0.04
Jun 18, 195	3	0.04	13.97	0	1.76	0.24	78.76	2.7
Jun 18, 195	4	0	16.18	0	0.9	0.11	78.56	1.16
Jun 18, 195	5	0	17.06	0.01	0.07	1.38	78.47	0
Jun 18, 195	6	0	9.69	0	0.02	17.61	70.9	0