Generic gasifier modelling: Evaluating model by gasifier type

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Abstract

There are many different types of gasifiers used for commercial or research purposes. These gasifier varieties differ in a number of ways, such as the direction of material flow or the physical chemical contact between the different species. These differences affect the modelling procedure and philosophy required to describe the gasification processes accurately.

Honeywell wishes to incorporate a generic gasifier model into their UNISIM DESIGN engine to simulate gasifiers accurately and calculate certain properties. Such a model does however not yet exist.

Presented in this document is a summary of the similarities and mutual properties among the different coal gasifiers, which allow for certain generic modelling procedures to be followed. The paper also highlights the discrepancies among these gasifiers and the areas where different modelling approaches should be followed. Apart from the specific gasifier characteristics, the phenomena of reaction kinetics, heat transfer and mass transfer were also investigated to ascertain their significance on gasifiers and specifically gasifier modelling. By following the guidelines provided in this paper, it should be possible to develop a generic gasifier model in any modelling environment.

KEYWORDS: gasifier, modelling, generic, summary, characteristics

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Nomenclature

$-r_A$	Rate of disappearance of species A per unit volume (gmol/s dm^3)
ΔT	Temperature difference between the surface and fluid (K)
$\frac{dT}{dx}$	Temperature gradient in the x direction (K/m)
ρ	Mass density of mixture (kg/m^3)
ρ_i	Mass density of species i (kg/m^3)
σ	Stefan-Boltzmann constant (5.676 × 10^{-8} W/m^2 K^4)
A	Pre-exponential factor
A_r	Area of the emitting surface (m^2)
Ar	Area normal to the direction of heat flow (m^2)
c	Total molar concentration (mol/m^3)
C_i	Concentration of species i ($gmol/dm^3$)
D_{AB}	Mass diffusivity of component A diffusing through component B (m^2/s)
E	Activation energy (J/mol)
F_{A_0}	Entering molar flow rate of species A (gmol/s)
F_A	Molar flow rate of species A (gmol/s)
h	Convective heat transfer coefficient (W/m^2)
J_A	Molar flux relative to the molar-average velocity (mol/m^2 s)
k	Thermal conductivity (W/m K)

k_A	Specific reaction rate
k_c	Convective mass transfer coefficient (mol/m^2 s mol/m^3)
N_A	Molar mass transfer of species A relative to fixed spatial coordinates (mol/s m^2)
q	Rate of convective heat-transfer (W)
q_r	Rate of radiant energy emission (W)
q_x	Heat-transfer rate in the x direction (W)
R	Gas constant (8.314 J/mol K)
r_A	Rate of generation of species A per unit volume (gmol A/s dm^3)
T	Temperature (K)
V	Volume of reactor (dm^3)
v	Mass-average velocity (m/s)
v_i	Velocity of species i (m/s)
y_A	Mole fraction of species A in the gas phase

Chapter 1

Introduction

Gasifiers are commonly used in chemical and petrochemical processes to extract energy from organic materials. Carbon monoxide (CO) and hydrogen (H_2) are usually the main desired effluent gases and have a large number of industrial and household applications. Gasifiers are not limited to the petrochemical industry, but find application in many fields. Different types of gasifiers are consequently required to fulfill the unique requirements.

Various researchers (Beychok (1974), Yoon et al. (1976), Yoon et al. (1978), Denn & Yu (1979), Kosky & Floess (1980), Arri & Amundson (1978), Britten (1988), Chen et al. (2000), Norman et al. (1997)) have developed system models (with differing levels of complexity) to describe the operation of gasifiers under specific operating and feed conditions. However, there is no universal, generically applicable model for coal gasifiers.

Honeywell is interested in developing and adding a generic gasifier unit to their UNISIM DESIGN engine, which will allow the accurate simulation of any specified type of gasifier.

This investigation was launched to gather information on as many different types of gasifiers as possible and to obtain the similarities and discrepancies among them from a modelling perspective. The derivation of a generic gasifier model was not undertaken yet, with the study rather focusing on comprehensive coverage of all the relevant material. This should provide the eventual developer of such a model with all the necessary information to set up the required models.

Chapter 2

Gasification

2.1 Overview

Gasification is the process whereby carbonaceous materials (petroleum, coal, biomass) are reacted with a specified quantity of oxygen at elevated temperatures (typically greater than 700℃). Gasification is a very effective way of extracting energy from organic materials, with the ensuing gas mixture commonly referred to as synthesis gas or just “syngas.”

Gasification differs from combustion in that all combustion processes are supplied with excess quantities of oxygen to ensure complete conversion of the feed. Complete oxidation of the feed is not attained in gasification, with gasification generally occurring in oxygen deprived environments. This results in partial oxidation of the feed. Gasification processes are however fed with a greater than stoichiometrically required quantity of oxygen to enhance the conversion to syngas. This oxygen quantity is however much less than that required for combustion, with as much as 50 % less oxygen fed than required for combustion. The amount of oxygen fed has an enormous effect on the eventual composition leaving the gasifier. This relationship is shown in figure 2.1.

When compared to gasification, combustion processes are typically much more exothermic. For fully converted combustion processes, nearly all of the feedstock’s chemical energy is released as heat, whereas in typical gasification, as much as 65 % of the energy is retained as so called “cold gas efficiency” and exits with the effluent as syngas.

Since gasification is so much less exothermic than combustion, it is vital to limit the heat transfer out of the gasification zone. This is required to ensure sufficient activation energy for the gasification reactions (at least 1000℃ is required to gasify coal).

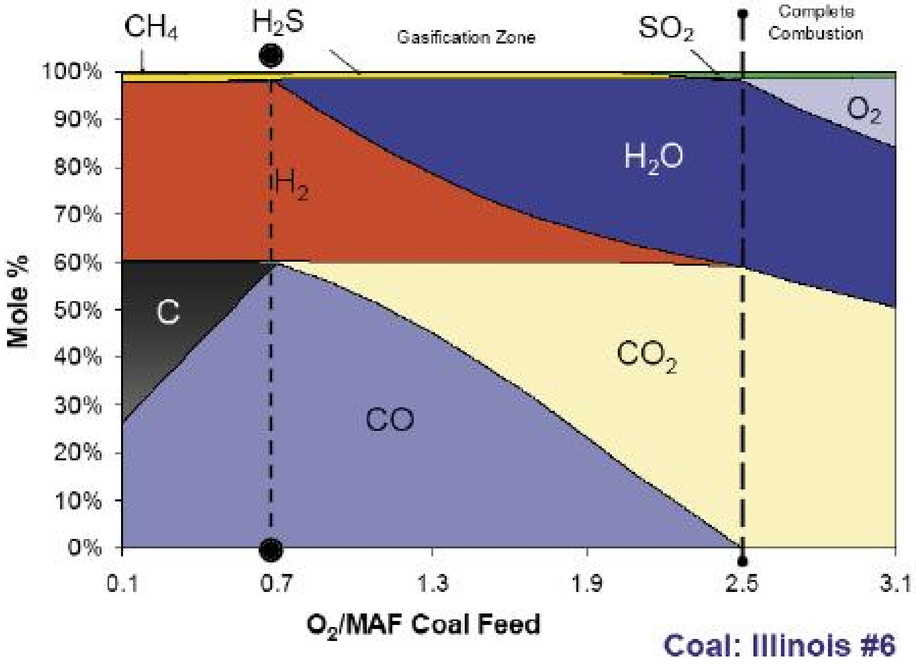

Figure 2.1: Product composition as a function of oxygen fed (Philips)

Gasifiers are therefore often refractory lined to prevent heat loss as much as possible, contrary to lining the reactor with water-filled tubes (as in combustion reactors) to absorb the heat.

2.2 Pyrolysis

As the carbonaceous particles heat up, the volatile material contained in the coal is removed via a process known as devolatilization. Char remains as by-product with an overall loss of almost 70 % of the coal mass. This process can also be described as "the chemical decomposition of a condenced substance through heating" (Anthony & Howard, 1976). The word "pyrolysis" is hence derived from the Greek words pyro – "fire" and lysys – "decomposition".

Pyrolysis is a sub-category of thermolysis[1] and occurs exclusively in organic matter. Severe cases, in which carbon is rendered as only residue is called carbonization and resembles charring. Pyrolysis is heavily dependent on the charecteristics of the organic substance and dictates the structure and composition of the char product, which ultimately takes part in the gasification reactions.

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[1] thermal decomposition

2.3 Products and reagents

The synthesis (product) gas produced during gasification is a fuel source itself, and is generally a more efficient fuel than the initial fuel fed to the gasifier. This is because more of the internal energy can be extracted from the synthesis gas than the original feed. The syngas can either be burned as is in internal combustion engines (producing hydrogen and methanol) or alternatively used as feed for the Fischer-Tropsch process and subsequently converted to synthetic fuels.

A distinct advantage of gasification is that low-grade fuels, such as organic waste and biomass can be used as raw material. In addition, almost any organic materials (even wood and plastic waste) can be used as feed to the gasification process. High temperature combustion has as advantage the rendering of cleaned gas production, in that corrosive ash elements (such as potassium and chlorine) are regularly refined out.

Oxygen gasifiers normally operate between 1250℃ and 1600℃, with air gasifiers not reaching such high temperatures, operating in the range 900℃ to 1000℃. The other obvious difference between the two types is the quantity of nitrogen and nitrogen related products associated with air gasification – these are absent in gasifiers using pure oxygen (Norman et al., 1997).

This study will focus solely on coal gasification, i.e. gasifiers using coal as feedstock.

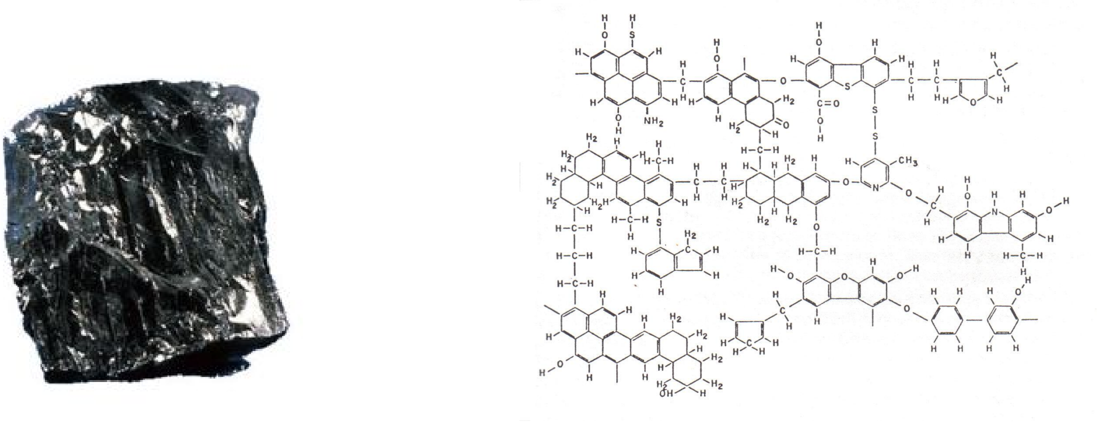
2.3.1 Coal

Coal is found all over the world, especially in the geologic column. Refer to figure 2.2 for a map showing the major coal fields of the world. Coal is formed through the metamorphosis of large quantities of plant residue by the effects of mainly pressure, but also temperature and time (Whitcomb & Morris, 1994). McMurry (2000) states that coal consists of many arrays resembling benzene-like rings attached together, which causes the coal to be a highly complex mixture. The result is a highly combustible blackish rock. See figure 2.3 for a picture of such a black coal rock as well as its chemical structure.

Coal is an important fossil fuel which consists of carbon, hydrogen and to a lesser extent sulfur and other trace elements. Coal is mined from underground or opencast coal mines. It is the largest fuel source for electricity production in the world as well as being a major contributor of carbon dioxide emissions.

Many different coal varieties are in use – the main difference among them being the time they were subjected to pressure. Listed in table 2.1 are the varieties as they transform from the lowest rank (on top) to the highest (at the bottom):

Figure 2.2: Coal map



(a) Sample of bituminous coal

(b) Chemical structure of coal

Figure 2.3: Coal (Nowacki, 1981)

Table 2.1: Types of coal

Type	Colour	Property	Use
Peat	–	precursor to coal	fuel in Scandanavia
Lignite	brown	lowest rank	fuel for electricity generation
Sub-bituminous	–	mixture between lignite and bituminous	fuel for steam electricity generation
Bituminous	black/dark brown	dense	fuel for steam electricity generation
Anthracite	black	highest rank	residential and commercial space heating
Graphite	–	difficult to ignite	pencils or lubricant

Coal Analysis

Coal fed to gasifiers undergoes devolatilization, a process whereby the chemical bonds of the coal are broken and the gases H_2, CO, CO_2, CH_4, H_2S, NH_3, water and some tars and oils are produced. A coal residue, char (predominantly fixed carbon) remains.

As a rule, different coals have different compositions and produce differing quantities of volatile components. A variety of coal gases can be produced from its thermal decomposition (Kosky & Floess, 1980). This is dependent on the conditions of pyrolysis. The pyrolysis process is complex and difficult to model (Anthony & Howard, 1976), so many researchers decide to select a devolatilization gas analysis, instead of attempting to predict the composition of the coal volatiles. Yoon et al. (1976) and Yoon et al. (1978) did extensive work on this topic and their results are widely used. Refer to table 2.2 for their suggested devolatilized coal composition.

Table 2.2: Composition of Devolatilized Coal

Component	Volume %
CO	20.6
CO_2	6.1
H_2	13.1
CH_4	50.3
H_2S, NH_3	9.9

The free swelling index is another often reported characteristic used to classify different coals. In essence the free swelling index is a measure of volatility and plasticity. Plasticity is vital for coking, as it represents the ability to form specific plasticity phases during the coking. This is measured by coal dilatation tests. Volatility is crucial for steel production and energy generation, as it determines the burn rate of the coal. Although high volatile coals are generally easier to ignite, they are often not as valuable as moderately volatile coals. Low volatility coals may be difficult to ignite, but will contain more energy per unit volume. Coking coal is therefore best if it has a very narrow range of volatility and plasticity.

Chapter 3

Common Gasification Processes

Gasification was initially developed to supply town gas for cooking and lighting during the early 1800s. Today it also finds industrial application to generate electricity. The development of the gasification process (in brief) was as follows (Higman & Van den Burgt, 2004):

1842: Baltimore Electric town gas

1887: Lurgi gasification patent

1925: Fischer-Tropsch patent awarded in Germany

1964-1965: An independent survey identified 65 gasifier types

1982: The subsequent survey narrowed the feasible field to 38

1983: Potential gasification processes feature 6 gasifiers

Today: Advanced gasification processes evolving

The type of gasifier preferred for a specific application depends on (among others):

- The required production rate of energy.
- The heating value of the gas.
- Operating temperature.
- Operating pressure.
- Desired gas purity/composition.

- Desired purity (presence of tars, ash).
- Availability of coal, as well as its type and cost.
- Constraints regarding size.

Gasifiers can be divided into four main categories:

1. Fixed bed gasifiers
2. Fluidized bed gasifiers
3. Entrained bed gasifiers, and
4. Molten bath gasifiers

3.1 Fixed-bed Gasifiers

In fixed bed gasifiers, steam and/or hydrogen are passed through large pieces of coal supported on a fixed grate. Fixed beds were traditionally unable to process coals with a tendency to cake[1], but recent modifications incorporate stirrers or rotating grates to avoid caking. Fixed bed gasifiers can be classified based on the direction of flow.

In fixed bed, counter-current ("up-draft") gasifiers, the gasification agent (oxygen and/or air and steam) flows through a fixed bed of carbonaceous fuel (coal or biomass) in a counter-current configuration, with the ash being removed as either slag or dry fly ash. For the slagging gasifiers to reach temperatures higher than the ash fusion temperature, a higher ratio of steam and oxygen to coal is required. In order to form a permeable bed, this type of gasifier requires a non-caking fuel with high mechanical strength, although recent developments have largely reduced the demands on the fuel specification. The throughput for these types of gasifiers is relatively low, but the lower gas exit temperature causes a higher thermal efficiency. This leads to increased production of tar and methane and a subsequent cleaning requirement of the product gas.

Co-current ("down-draft") fixed bed gasifiers, are very similar to the counter-current types, with the exception that the gasification agent flows co-currently with the fuel (i.e. in a downward configuration). Heat is added to the top part of the bed either using external heat sources, or through the combustion of small quantities of the

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[1] a process whereby the coal agglomerates to form a cake when heated

fuel. The product gas therefore exits the gasifier at an elevated temperature, with the majority of this heat being transferred to the gasification agent in the upper bed. This yields an energy efficiency on par with the counter-current configuration. Tar levels are significantly lower than in counter-current gasifiers, as all the tars pass through a hot char bed.

In addition to classification based on flow direction, fixed-bed gasifiers can be further divided into single- or two stage units. The primary difference is the location of gas removal, although the temperature ranges attained inside the drying and devolatilization sections also differ. Single stage gasifiers are associated with a single gas offtake above the drying zone, with effluent temperatures generally between 370℃ and 595℃. This causes the entering coal to be heated rapidly.

Two stage gasifiers have two product gas offtakes – one above the drying zone (similar to single stage designs) and another above the gasification zone. Approximately 50 % of the gas formed during gasification is removed at this second offtake, while the remaining gases are transported upwards through both the devolitilization and drying sections. The temperatures attained in two stage gasifiers are significantly lower than those of single stage gasifiers.

The fixed-bed gasifier systems that have contributed most to the development of the contemporary fixed-bed gasifier are discussed below.

3.1.1 British Gas/Lurgi Slagging Process

Project Development

1953: Lurgi experimented using a slagging variation of their fixed-bed gasifier.

1955: The Ministry of Power acquired Lurgi's technology. They subsequently built and operated a pilot version at the Gas Council's Midlands Research Station (MRS) until 1958.

1960: The Wilson Committee proposed that the development of Lurgi's slagging gasifier should be carried on under the guidance of the Gas Council. The MRS gasifier was significantly altered to operate at 26 atm.

1974: The British Gas Corporation begins to adapt a Lurgi gasifier to accommodate slagging. Alterations were finalised by 1975 and the first run commenced shortly after.

1977: Conoco initiates a demonstration plant that would be able to convert bituminous coal into pipeline quality gas.

Technology

The Gas Council's initial slagging gasifier had a vertical orientation, was cylindrical, jacketed (water based) and operated at approximately 5 atm. A lock hopper at the top was used to feed the coal feedstock, while steam and oxygen were premixed and fed through tuyeres[2] in the gasifier outer wall. The slag was removed sporadically using a tap on the side of the gasifier.

The 1962 edition could operate at a higher 26 atm, while the slag tap was moved to the bottom of the gasifier. The gasifier built in Leatherhead (BCURA), had an operating pressure of 3 atm and slag was removed continuously.

A further improvement was introduced at the Westfield site in Scotland, where the slag tap was fitted with a burner to ensure a free flow of slag. Refer to figure 3.1 for a detailed schematic of the British Gas/Lurgi Slagging gasifier.

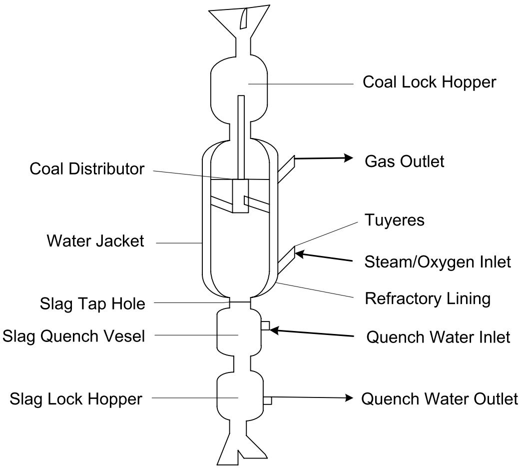

Figure 3.1: British Gas/Lurgi Slagging gasifier (adapted from Hartman et al. (1978))

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[2] a tube, nozzle or pipe through which air is blown into a furnace or hearth (Gale, 1972)

Operating Conditions

Temperature:

- Combustion zone: 1260°C to 1370°C
- Effluent gas: 370°C to 425°C

Pressure: 5 atm to 26 atm

Residence time:

- Gas velocity: roughly 0.75 m/s above the slag bath
- Coal residence time: 10 min to 15 min

Reactants

Coal: Bituminous caking coals accepted with refractory ash up to 15 % wt and 20 % wt moisture. Fines smaller than 1/8" diameter removed and injected with steam and oxygen.

Oxidant: Successful gasification requires approximately 0.52 ton oxygen per ton of coal.

Steam: Successful gasification requires approximately 0.28 ton steam per ton of coal.

3.1.2 GFERC Slagging Process

Project Development

The GFERC slagging gasifier was designed as an improvement on the Lurgi fixed bed, dry ash gasifier.

These dry ash gasifiers were developed during the 1930s and consisted of a bottom grate for ash disposal. Excess steam was also fed to keep the operating temperature low enough to form dry ash.

1958-65: The Bureau of Mines (Department of Interior) designs and operates the slagging gasifier pilot plant at GFERC. This was mainly done to ascertain the feasibility of slagging operations and to evaluate operating parameters.

1965-74: The pilot plant was mothballed.

1976: The increased interest in gasification of coal led to the re-startup of the GFERC process.

Technology

Figure 3.2 is a schematic of the GFERC gasifier. The height of the fuel bed can be adjusted by removing some or all of the intermediate sections.

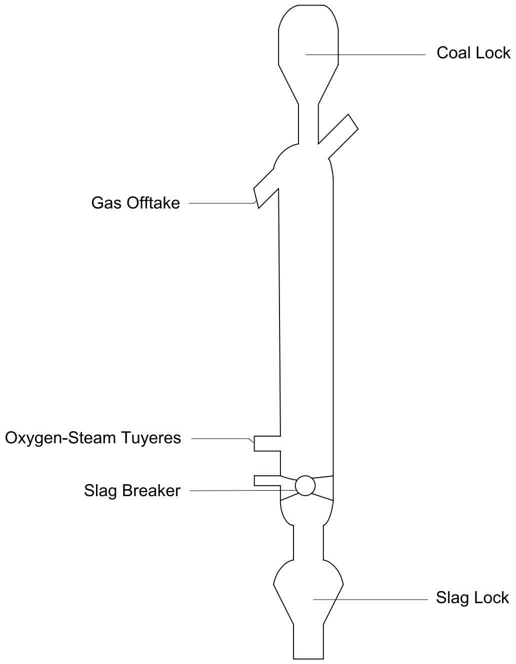


Figure 3.2: GFERC Slagging gasifier (adapted from Gronhovd & Kube (1977))

3.1.3 Lurgi, Dry Ash Process

Project Development

1936: The first full scale Lurgi coal gasifier built in Hirschfelde, Germany.

1936-present: More than 18 commercial plants established around the world. One of the most notable is the Sasol plant in South Africa which uses more than 16 gasifiers to produce synthesis gas for synthetic fuels production.

Technology

The fixed bed Lurgi gasifier is a vertical, cylindrical reactor vessel (figure 3.3).

Lurgi gasifiers are usually made up of three sections: the coal bunker and air lock at the top, followed by the gasification chamber and finally the ash lock at the bottom. A conveyor belt is typically used to transport the coal into the coal bunker. From here the coal is pressurized before being fed into the gasification chamber. Hydraulic valves are used to seal and open or close the coal bunker (De ponte et al., 2001).

The gasification chamber consists of a double layer, with the void between the two walls used to produce steam. Water is heated by the heat of the gasification reactions and the ensuing steam used as reagent in the gasification chamber. Another important component is the rotating grate. The grate rotates at variable speeds to regulate the amount of ash removed. The grate also breaks apart the bigger coal lumps to increase the surface area and prevent blockage. By regulating the amount of ash removed, a handle is obtained with which to control the height of the reaction bed in the gasification chamber.

The ash lock operates in a similar fashion to the coal lock, in that two hydraulic valves are once again used to regulate the depressurization and removal of the ash.

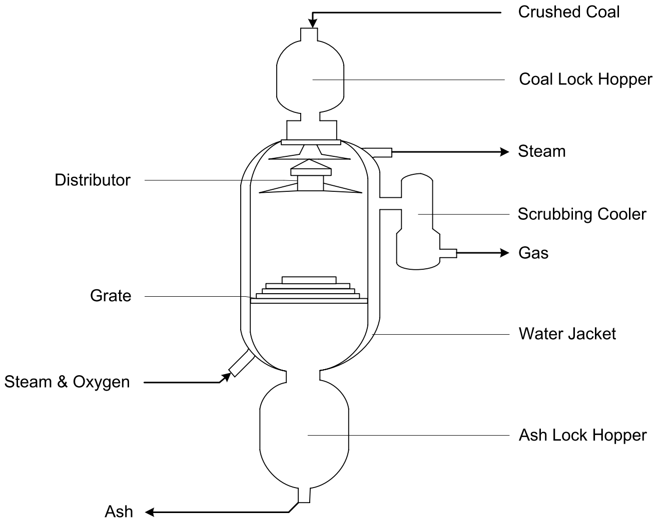


Figure 3.3: Lurgi, Dry Ash gasifier (adapted from Hartman et al. (1978))

Operating Conditions

Temperature:

- The temperature depends on the type and make-up of feed coal, but falls in the following general ranges:
- Gasification zone: 620℃ to 815℃
- Combustion zone: 980℃ to 1370℃
- Effluent gas: 370℃ to 590℃

Pressure: 2.4 MPa to 3.1 MPa

Coal residence time: up to 1 hour

Reactants

Coal: The dry ash gasifier requires non-caking coals, except if a mechanical stirrer is provided.

Oxidant: Can be either air or oxygen blown. Oxidant quantity is dependent on coal type, but typically 0.6 ton oxygen is used per ton of coal.

Steam: Steam quantity is dependent on the type of coal, as well as oxygen consumption. 3.2 tons of steam per ton of coal is typically required.

3.1.4 Wellman-Galusha Process

Project Development

1896: Wellman Engineering Company manufactures gasifiers. These early gasifiers have anthracite, lignite, coke, charcoal as well as bituminous coal as feedstock and operate at atmospheric pressure.

1968: A new company, McDowell-Wellman Company is formed and commences development on a pressurized alternative to the atmospheric gasifier. The project was funded by the Office of Coal Research.

Technology

There are standard and agitated varieties of the Wellman-Galusha gasifier. The agitated gasifier differs in that it has a larger (almost 25 %) rated capacity than the standard type of similar size, as well as being able to process volatile caking bituminous feed coals (figure 3.4).

The gasifiers are also water jacketed, with the entire gasifier being enveloped.

The agitator of the agitated gasifiers consists of a horizontal rotating arm. This arm is also used to preserve a consistent fuel bed and impede channeling by spiraling vertically below the coal bed surface. The agitator's position and velocity can also be changed to accommodate varying feeds and operating modes.

The gasifiers are also fitted with a rotating step grate on a center post at the bottom of the gasifier. It dispenses the air and steam into the coal bed as well as driving the ash into the ash cone.

A coal bin at the top of the gasifier is used to house the sized coal, from where it is gravity fed into the feeding section. The flow into the feeding section is regulated by two slide valves which are normally closed, and only opened momentarily when refilling. The feeding section then supplies coal to the gasifier on a continuous basis through vertical feed pipes, again with gravity as driving force. Two more slide valves are situated at this outflow. These valves on the contrary are usually open, but closed during refilling. The continuous inflow of coal permits the stabilization of the coal bed and gas quality – which makes this a highly favoured sequence.

The air required for gasification is fan-blown across the water jacket, allowing steam to be absorbed and carried along. The amount of steam taken up is controlled by regulating the temperature of the water jacket.

Operating Conditions

Temperature:

- Combustion zone: 1315°C
- Effluent gas: 590°C to 650°C, for bituminous coal

Pressure: atmospheric

Residence time:

- Gas velocity: Fluctuates depending on position in fuel bed.
- Coal residence time: 4 hours

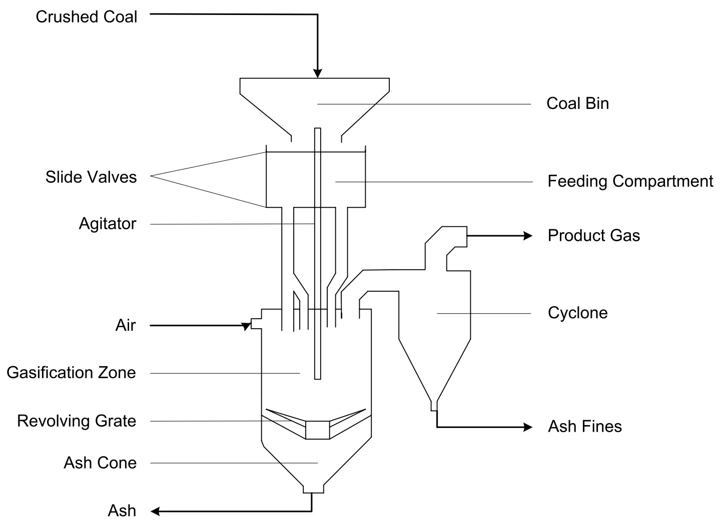


Figure 3.4: Wellman-Galusha gasifier (adapted from Hartman et al. (1978))

Reactants

Coal: The standard gasifier can process anthracite, coke and charcoal, whereas the agitated type can also handle bituminous and sub-bituminous coals, and has no limit on the free-swelling index of coals to be used.

Oxidant: Air is used and provided at approximately 3.5 tons per ton of coal.

Steam: Supplied from the cooling water at between 0.4 and 0.7 tons per ton of coal fed.

3.1.5 Woodall-Duckham/Gas Integrale Process

Project Development

1940: II Gas Integrale develops a two-stage gasification process in Milan, Italy. The gasifier is used in a cyclic process.

Process licensor Babcock Contractors, Inc. takes over the project as the Woodall-Duckham/Gas Integrale process after 20 years of cyclic operation.

Technology

Figure 3.5 shows a schematic of the Woodall-Duckham/Gas Integrale gasifier.

It is a cylindrical, vertical reactor. It consists of a concentric ring, rotating grate situated in the bottom of the vessel. The rotating grate serves to distribute the inlet air and steam and dispose of the ash formed during reaction.

The gasifier can be divided into two zones – a refractory lined drying and distillation zone and a water-jacketed gasification zone.

Coal is fed to the gasifier through the lock-and-surge hopper system. From here it enters the flooded coal distributer. The first zone encountered is the drying zone, where the water content is depleted by counter-current contact with the hot gases rising from the gasification zone. The temperature of this zone is about 120℃.

The distillation zone follows the drying zone. This zone is slightly hotter than the drying zone and causes the volatile matter contained in the coal to distill into the rising gas stream. In the case of caking coals, fractional fluidization will occur, with the coal subsequently resolidifying to form semicoke.

The gasification zone operates close to the gas fusion temperature. Here the residual carbon is gasified as it is contacted counter-currently with preheated air and steam in an unagitated fixed bed.

The water jacket is primarily used to produce steam. The air to steam ratio is regulated to ensure a gritty ash, as was found that a gritty ash with a number of small clinkers is much easier to handle. Low steam contents cause higher reaction temperatures which lead to large clinker production. The opposite is true for high steam contents and the result is a very fluffy ash.

Another interesting feature of these gasifiers is the two gas-outlet positions. The first outlet is positioned just above the gasification zone, with the other found at the top of the gasifier above the drying zone. The first gas withdrawn is called clear gas. It is used to regulate the temperature in the distillation zone. Decreasing the clear gas flow causes more hot gas to be fed to the distillation zone, increasing its temperature.

Operating Conditions

Temperature:

- Gasification zone: 1200℃
- Clear gas (bottom offtake): 650℃
- Effluent gas (top offtake): 120℃

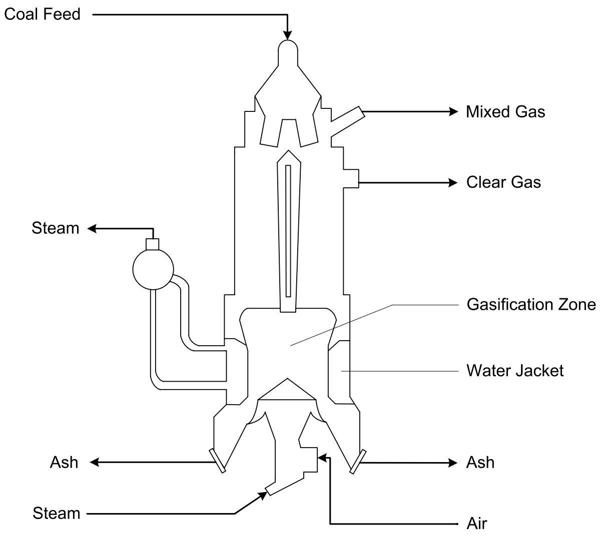


Figure 3.5: Woodall-Duckham/Gas Integrale (adapted from Hartman et al. (1978))

Pressure: atmospheric

Residence time:

- Gas velocity:
  - Gasification zone: Equivalent to one-stage gasifiers
  - Devolitilization/drying zone: Lower velocity causes reduced carryover of fines.
- Coal residence time: several hours

Reactants

Coal: Double screened. Coals accepted with free-swelling index < 2.5.

Oxidant: Air provided at 2.3 ton per ton of coal.

Steam: Steam produced internally at a rate of 0.25 ton per ton of coal.

3.2 Fluidized Bed Gasifiers

In fluidized bed gasifiers, the reagent gases are passed through finely ground coal particles causing a boiling and elevating effect. This yields a higher bed with greater coal surface area. Fluidized beds are not suited to caking coals. The caking attributes of these coals can however be reduced by pre-treatment. Oxygen or air and steam is used to fluidize the fuel mixture, while the ash is removed either dry or as defluidized heavy agglomerates. The fuel in dry ash gasifiers should be highly reactive, as the associated temperatures are generally low. Low-grade coals are usually sufficient. The temperatures achieved in the agglomerating gasifiers are slightly higher, so better grade coals are used. Fuel throughput in fluidized bed gasifiers is not as high as in entrained flow gasifiers, but higher than for the fixed bed alternative. The presence of carbonaceous materials and their subsequent elutriation causes the conversion efficiency to be fairly low. The efficiency can be increased by recycling or combustion of these solids. Fuels that regularly form highly corrosive ash (such as biomass fuels) and subsequently damage the sides of slagging gasifiers, are normally used in fluidized bed gasifiers.

Several fluidized bed gasifiers have contributed largely to the development of fluidized bed gasifier technology. This report will focus only on a selected few of these.

3.2.1 Battelle Ash Agglomerating Process

Project Development

Early 1960s: Union Carbide Corporation finances the research of the gasification process at Battelle for a Power Distribution Unit (PDU). The research focused on the performance of agglomerating ash burners in bench scale vessels using both bituminous and sub-bituminous coal. Experiments were also conducted to ascertain the influence of erosion on turbine blades caused by fly ash from the agglomerating ash conditions.

1965: Both Union Carbide and Battelle begin their pursuit to interest the Office of Coal Research (OCR) in the development of the project.

1972: OCR and the American Gas Assosiation (AGA) agree to sponsor subsequent work in collaboration. The AGA authorizes the design and construction of a unit to operate under pressure.

1973: Installation of the pressurized vessel commences.

1974: The AGA withdraws its subsidy. The project was henceforth solely supported by the OCR. Field construction of the PDU in West Jefferson, Ohio begins.

1976: PDU construction completed.

1978: PDU shut down.

Technology

The Battelle process makes use of two different vessels: the burner and the gasifier, which are both refractory lined.

The gasifier is of vertical, cylindrical construction and consists of three distinct sections, each with a different diameter, increasing upwards. See figure 3.6.

Inert gas is used to transport coal pneumatically to lock hoppers. These lock hoppers are kept at a higher pressure than that of the gasifier system and serves both the burner and gasifier. Recycled product gas is employed to convey the coal from the lock hopper into the gasifier.

The burner is fed with coal (carried by air), recycled char and ash from the gasifier as well as additional air. The coal, char and ash blend is fluidized as combustion takes place.

As oxidation of the coal and char occurs, ash particles combine and overflow the burner, from where they enter the gasifier by means of a steam lift. Coal enters the gasifier through a nozzle above the ash entrance. Superheated steam enters the gasifier through a distributor plate at the bottom and hence provides fluidization.

The char and ash particles are segregated based on different superficial gas velocities and densities. The two zones are separated by an inner layer consisting of ash agglomerates, char and coal. The bottom zone contains fluidized ash agglomerates, while the top layer contains mainly fluidized char and extends to the char withdrawal point.

The heat transfer between the carbonaceous material and agglomerates provides the endothermic reaction heat. Gasification of the coal is limited, with the resulting char and coal being extracted and fed to the burner.

Operating Conditions

Temperature:

- Burner: 1100℃ to 1150℃
- Gasifier: 870℃ to 890℃
- Effluent gas: 870℃ to 890℃

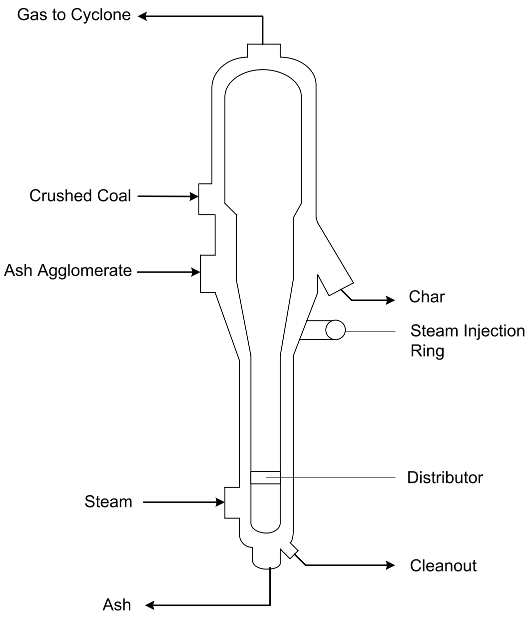


Figure 3.6: Battelle Ash Agglomerating Gasifier (adapted from Hartman et al. (1978))

Pressure:

- Pretreater: Atmospheric
- Burner: 690 kPa to 860 kPa
- Gasifier: 690 kPa to 860 kPa

Reactants

Coal: Crushed to -8 +100 mesh. Accepts only non-caking coals, except if pretreatment section is provided. Ash fusion temperature not critical.

Oxidant: Air used at rate of approximately 10 % to 15 % more than stoichiometrically required for combustion.

Steam: Successful gasification requires between 0.8 and 1.2 ton steam per ton of coal.

3.2.2 Carbon Dioxide Acceptor Process

Project Development

1968: The Carbon Dioxide Acceptor prototype together with feasibility studies completed and approved. Rapid City, South Dakota is selected as the preferred location for the Conoco Coal Development Company's (CCDC) Carbon Dioxide Acceptor process. The project is funded equally by the DOE and AGA.

1971: Erection of most of the pilot plant facility completed. Only the methanation component to complete.

1972: Operation of the pilot plant commences.

1974: The methanation component completed.

Laboratory work continued throughout to improve as well as resolve some of the remaining issues.

1976: Environmental analysis launched by the Radian Corporation. Commercial design study concluded.

Technology

Lignite or sub-bituminous coal is fed from a lock hopper to the bottom of the fluidized bed gasifier (figure 3.7). Before entering the lock hopper, the coal is diminished to +100 -8 mesh, pre-heated and dried to a water content of less than 5 %.

Steam is also introduced from the bottom, while the acceptor (either calcinated limestone or dolomite) is injected from a regenerator at the top of the fluidized bed.

The regenerator is fed with solid dolomite or limestone (external), as well as char, air and steam from the gasifier. It evolves carbon dioxide and discharges chemical energy to the gasifier. Char is combusted to supply the heat required to regenerate the acceptor.

The coal in the gasifier is subsequently devolatilized and gasified in the company of steam, the acceptor, carbon monoxide and hydrogen.

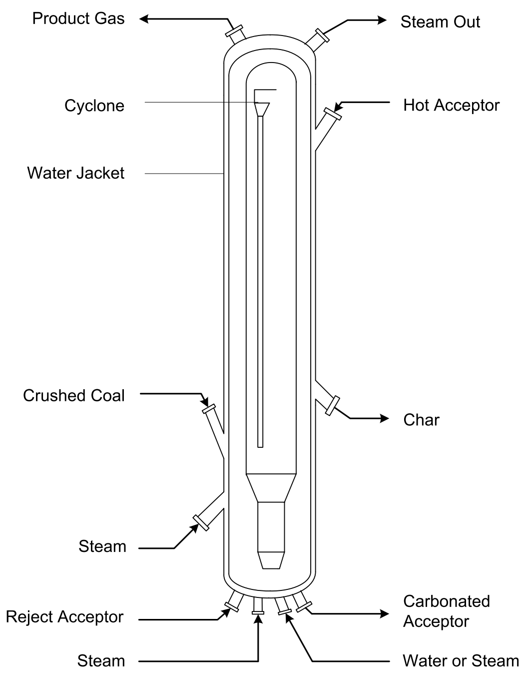


Figure 3.7: Carbon Dioxide Acceptor Gasifier (adapted from Hartman et al. (1978))

Operating Conditions

Temperature:

- Gasifier: 815℃
- Regenerator: 1000℃

Pressure: 1030 kPa

Residence time:

- Gas velocity:
  - Empty gasifier: 0.5 m/s to 0.7 m/s
  - Fluidizing velocity: 0.35 m/s
  - Regenerator: 0.9 m/s (fluidizing)

Reactants

Coal: Lignite or sub-bituminous is dried to 5 % moisture and crushed to -8 +100 mesh.

Oxidant: Air not required for gasifier, but rather fed to regenerator at 2.3 tons per ton of lignite coal.

Steam: Successful gasification requires 1.1 tons steam per ton of coal.

3.2.3 HYGAS Process

Project Development

1946: The joint operation between the Institute of Gas Technology and the DOE, HYGAS, commences.

1971: A pilot plant is erected in Chicago, Illinois.

1973: Pipeline quality gas produced from coal on a sustainable basis.

1974: Successful integration with steam/oxygen gasification accomplished. During this year a two megawatt electrothermal gasifier was also investigated. Although it supplied a desirable rich hydrogen-gas and was proved to be technically feasible, the project was abandoned as the electrical costs were too great.

1975: Tests are conducted to ascertain the gasifier's suitability to process different types of coal. Most coals are found to be processed adequately.

1976: Research aimed at determining effects of acidic condensing atmospheres on insulation as well as evaluation of methanation catalyst. These tests were concluded in December 1976.

1978: A wider variety of coals are tested. Tests on different pressure regimes (up to 3400 kPa) were also conducted.

1979: Further modifications are made which include studying the relationship between gas velocity and pressure drop. Further tests are conducted to improve the carbon conversion at current feed rates, minimize the requirement on external pre-treatment and to employ the recycled coal fines from a reactor system.

Technology

The HYGAS gasifier consists of four internal gas/solid contacting sections (figure 3.8). These stages are interconnected through transfer lines and are (from top to bottom):

- Slurry drying/solids extrication

- 1st stage Hydrogasification
- 2nd stage Hydrogasification
- Gasification

Coal is slurried with a light oil by-product and injected at the top of the slurry drying section where the oil is removed by vapourization. The oil exits along with the raw gas and is retrieved for reuse.

In the first hydrogasification zone, the carbon-hydrogen reaction takes preference to yield excess hydrogen. Coal particles from the drying zone descend through a lift pipe (gravity fed) to contact the hot gases from the zone below counter-currently. This lift pipe serves as the first hydrogasification zone. The gas also serves to transport the remaining solids to the gas/solid extrication zone. Temperatures in this section are typically between 650℃ and 700℃.

The second hydrogasification zone follows and houses the contact between the descending partially gasified char and the hydrogen rich gas from the gasification section below. Almost a quarter of the feed coal is converted in this section (while between 45 % and 50 % of the total carbon convertion occurs in the two hydrogasification sections). This section has the most dense fluidization with temperatures from 920℃ to 980℃. A rise in temperature speeds up the endothermic reactions, while a drop subsequently slows it down.

The steam/gasification section accepts the residual coal form the above sections together with steam and oxygen from below. The excess hydrogen reacts with carbon and carbon monoxide to form methane. The exothermic nature of these reactions supplies much of the energy needed by the endothermic steam-carbon reaction. The oxygen consumption is therefore reduced. As a result, this process requires the least oxygen of all oxygen based gasifiers surveyed.

Operating Conditions

Temperature:

- Slurry dryer zone: 315℃
- 1st Hydrogasification zone: 540℃ to 690℃
- 2nd Hydrogasification zone: 940℃
- Gasification zone: 1000℃

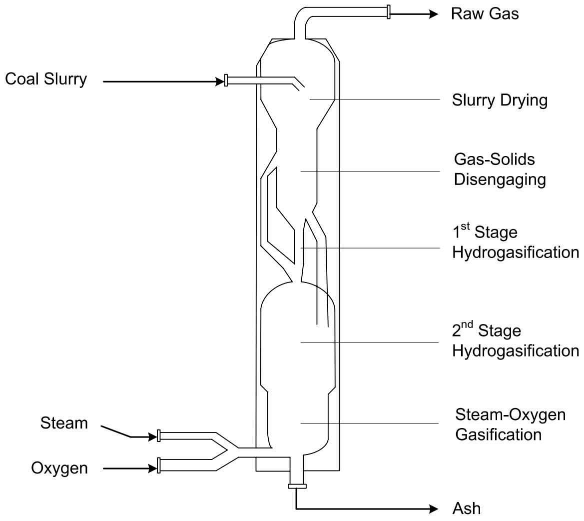


Figure 3.8: HYGAS Gasifier (adapted from Hartman et al. (1978))

Pressure:

- Slurry dryer zone: 7960 kPa
- 1st Hydrogasification zone: 8000 kPa
- 2nd Hydrogasification zone: 8030 kPa
- Gasification zone: 8100 kPa

Reactants

Coal: Any rank, moisture or sulfur content can be used. Coals pulverised to -10 +100 mesh.

Oxidant: Oxygen required at 0.22 to 0.25 ton oxygen per ton of sub-bituminous coal and 0.24 to 0.26 ton per ton of bituminous coal.

Steam: Successful gasification requires 1.0 to 1.2 tons of steam per ton of coal, regardless of the type of coal.

3.2.4 Synthane Process

Project Development

1961: Efforts began to improve the methods for pretreating caking coals and high pressure operation in fluidized bed gasifiers.

1970: The U.S. Bureau of Mines develops the Synthane process and awards the initial design contract to M.W. Kellogg Company.

1971: The contract for the design of the pilot plant is awarded to Rust Engineering. It was subsequently built in Bruceton, Pennsylvania.

1975: Plant construction completed.

1976: Design changes suggested and implemented.

1978: Testing extended to include the use of Illinois No. 6 coal (a highly caking coal) and successful pre-treatment and gasification achieved.

1979-80: A lack of funding necessitates the premature shutdown of the gasifier – it was subsequently put on standby for two years.

Technology

The two novel features of Synthane gasifiers are that the pre-treatment of caking coals is combined with gasification, and that the product gas consists of a high methane content.

Coal (crushed to -20 mesh) is transported into the fluidized bed by high pressure oxygen and steam. Before entering the gasifier, the coal is dried and pressurised to about 40 atmospheres in a pre-treater. The pre-treatment prevents the agglomeration of caking coals by slightly oxidizing the coal surface.

Steam and oxygen enter the fluidized bed through an inlet below the fluidizing gas distributor. A schematic of the Synthane gasifier can be seen in figure 3.9.

The char contacts the steam counter-currently as it flows downwards in the fluidized bed. The residual char is removed through a let down valve.

The product gas exits at the top and passes through a series of scrubbers to remove ash and other impurities entrained in the gas.

Operating Conditions

Temperature:

- Fluidized bed: 815℃ to 980℃
- Effluent gas: 425℃ to 870℃

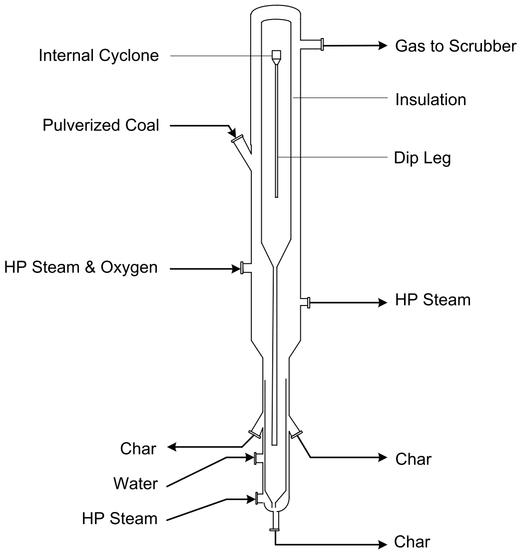


Figure 3.9: Synthane Gasifier (adapted from Hartman et al. (1978))

Pressure: 4135 kPa to 6900 kPa

Linear gas velocity: 0.1 m/s to 0.2 m/s

Reactants

Coal: All types of coal are accepted, although caking coals require pre-treatment. Moisture content should be below 14 % (mole basis) and coal pulverised to 20 × 0 mesh.

Oxidant: Oxygen required at 0.35 ton oxygen per ton of caking coal. Non-caking coals require less oxygen.

Steam: Successful gasification requires approximately 1.5 tons of steam per ton of caking coal. Non-caking coals require less steam.

3.2.5 Winkler Process

Project Development

1920: The need to produce industrial fuel- and synthesis gas from coal on a commercial scale arises. The Winkler process is developed and preliminary work commences.

1926: The first commercial Winkler gasifier begins operation in Leuna, Germany. During the late 1920's tests were conducted on soft U.S. coals. The gasifier proved to process these adequately.

1938-73: Davy Powergas installs and operates more than 16 Winkler gasifiers throughout Western and Eastern Europe as well as the Far East. Initial gasifiers operated atmospherically and with air as oxidant. Throughput was increased by increasing the pressure, and further increased by switching to oxygen as oxidant.

1983-84: The first four trains of a high temperature Winkler gasifier built near Cologne, Germany.

1987: Methanol plant completed.

Technology

The Winkler process consists of a cylindrical, vertical, refractory lined gasifier (figure 3.10). The coal is introduced in the bottom of the gasifier, while the gas (steam and oxygen/air) enters via nozzles fitted at various places along the gasifier. Their primary inlet is at the bottom, while the higher nozzles serve to gasify the unconverted coal exiting the fluidized bed. The coal is fed from feed bunkers using variable speed screws, but first passes through a series of crushers and driers to prepare the coal for gasification. The feed screws maintain the feed rate of the coal adequately.

The fluidized bed fills approximately the lower third of the gasifier's capacity. The remainder of the volume is used as a disengagement zone. The fluidized bed is kept at a high temperature, ensuring that all the heavy hydrocarbons and tars are gasified as well.

Segregation of the ash particles in the fluidized bed occurs based on size and density differences. The heavier ash particles trickle down and are captured in the ash discharge chamber.

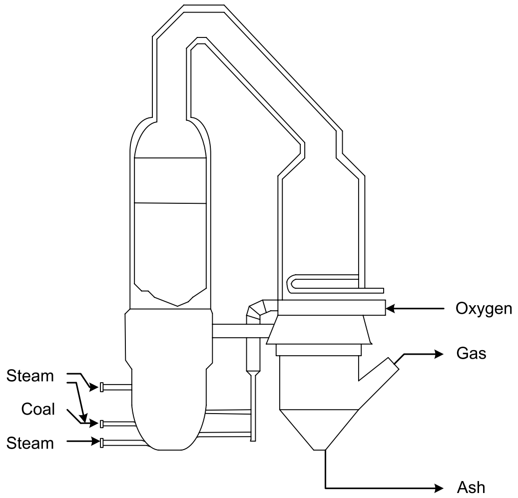


Figure 3.10: Winkler Gasifier (adapted from Hartman et al. (1978))

Operating Conditions

Temperature:

- Fluidized bed: 980℃ to 1150℃
- Effluent gas: 790℃ to 1150℃

Pressure: 100 kPa to 400 kPa

Coal Residence Time: 20 minutes to 30 minutes

Reactants

Coal: Crushed coal is necessary. Lignite or sub-bituminous coal with moisture content less than 18 % (mole basis) is preferred. Ash content of coal not crucial – as high as 50 % reported to have been processed successfully.

Oxidant: Oxygen or air. Oxygen required at 0.35 to 0.6 ton oxygen per ton of coal. Air at 1.7 to 3.0 ton per ton of coal.

Steam: Successful gasification requires approximately 0.4 to 0.7 tons of steam per ton of coal (for oxygen blown gasification).

3.3 Entrained Bed Gasifiers

Entrained bed gasifiers also employ fine coal particles, which are carried into the gasifier by the reagent gases. Any type of coal can normally be used in these systems. Oxygen is used to gasify either a dry pulverized solid, a fuel slurry or an atomized liquid fuel. This happens in a co-current manner. The oxygen is sometimes replaced with air. Due to the high operating temperatures, almost any type of coal can be fed to these gasifiers. The coal particles are also well separated from one other, with the gasification reactions taking place in a dense, fine cloud of particles. Because of the high temperatures and pressures (gasifiers usually operate at pressures as high as 6 MPa) attained, a higher than normal throughput is achieved, albeit at the expense of lower thermal efficiency. This is because the gas has to be cooled down before the current technology is able to clean the gas.

Another advantage of the higher temperature is the absence of both methane and tar in the product gas make-up. However, the oxygen requirement is higher than the other types of gasifiers. The operating temperature is also higher than the ash fusion temperature, causing the majority of the ash to be removed as a slag. The rest of the ash is either a blackish fly-ash slurry or very fine dry fly-ash. The slag formed from certain fuels (particularly biomass) can be very corrosive to the ceramic inner gasifier walls. Their major function is to protect the gasifier outer wall. To counteract this, some gasifiers are lined with a water cooled wall covered with partially solidified slag.

Certain fuels lead to ashes with extremely high fusion temperatures, but these can be lowered by adding limestone to the fuel. Entrained bed gasifiers require the smallest fuel particles of all the industrial types of gasifiers. Their fuels are therefore pulverized first, which adds to the already high energy consumption of these gasifiers. The production of the oxygen required for entrained bed gasifiers is however a far greater contributer to the overall energy burden.

The most significant and widely available entrained bed gasifiers are discussed below.

3.3.1 Babcock and Wilcox Process

Project Development

1951: Babcock and Wilcox Company built their first pilot gasifier at the Bureau of Mines Station in Morgantown, West Virginia. The gasifier had a capacity of only 6 tons of coal per day. Later that year, the Du Pont Company erected a

Babcock and Wilcox gasifier at their plant in West Virginia – it's capacity was six times greater than that of the original gasifier.

1955: Du Pont builds an even larger gasifier (capacity 400 tons per day). When natural gas became abundant, the gasifier was modified to run on partly oxidized natural gas.

1960: Babcock and Wilcox erects a gasifier in Alliance, Ohio to operate using air as oxidant. Another gasifier was constructed in Morgantown which operated at an increased pressure of 2100 kPa (up to then all of these gasifiers operated at atmospheric pressure). A down fired axial coal feed was also added.

Technology

The Babcock and Wilcox gasifier is a cylindrical, vertical, double shelled vessel, with the outer shell housing the refractory (figure 3.11). Two horizontal rows of burners are situated near the bottom of the gasifier.

The coal is kept in a feed tank in a pre-pulverised and dried-out state. The coal is injected into the coal burners of the gasifier together with air and oxygen. Burners of different types are used to negate pre-mixing of the reactants and subsequently prevent flashback.

The operating temperature of these gasifiers is significantly higher than the ash fluid temperature. This causes the ash to form a molten slag, which is continually removed into a molten slag tank. The slag is quenched and released to slag lock hoppers from where it is disposed.

The unreacted char exits the gasifier along with the product gas, is captured in a system of cyclones and recycled back to the gasifier reaction zone.

Operating Conditions

Temperature:

- Combustion zone: 1870℃
- Effluent gas: 980℃

Pressure: 100 kPa to 2000 kPa

Gas velocity residence time: vertical velocity between 4.5 m/s and 9 m/s, with residence time approximately 1 second.

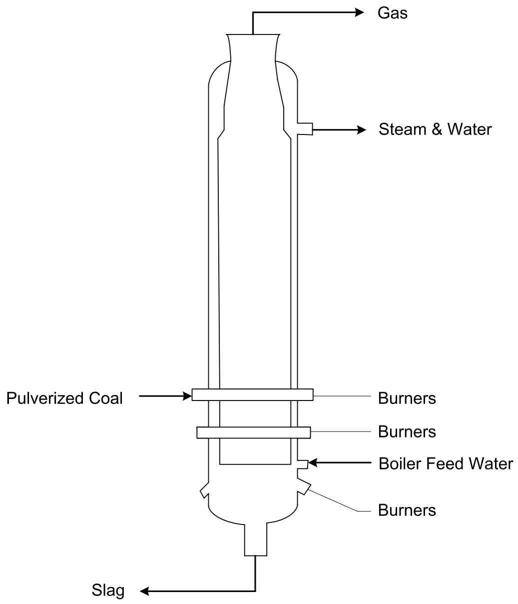


Figure 3.11: Babcock and Wilcox Gasifier (adapted from Hartman et al. (1978))

Reactants

Coal: Accepts most coal types, pulverised to 70 %, 200 mesh. Moisture content is not critical but affects pulverised coal flow.

Oxidant: Oxygen required at 0.8 to 1.0 ton per ton of coal.

Steam: No steam required. Water used for temperature control – 0.05 ton per ton of coal.

3.3.2 BI-GAS Process

Project Development

1963-74: Bituminous Coal Research Inc. (BCR) started the program "Gas Generator Research and Development" with primary goal to produce a high energy content pipeline gas from coal, using the BI-GAS process. Initial experimentation showed that a highly methane rich gas can be produced by reacting coal with steam at high pressures and temperatures. The next two phases involved operating continuously and testing various types of coal. During further investigations, it was found that the physical design of stage 2 (figure 3.12), affected the methane formation. Techniques to improve the flow patterns in stage 2 were pursued to ultimately propose design criteria for the slagging part of the gasifier.

1974: BCR completed the fundamental design criteria for the BI-GAS process and a pilot plant management contract was awarded to Phillips Petroleum Company.

1976: The pilot plant was completed and operation commenced. BCR continued their research and investigated the differences between continuous and semi-continuous modes.

1978: Slag removal posed numerous difficulties which lead to many unscheduled shutdowns.

1979: Stearns-Roger took over the project.

Technology

The BI-GAS gasifier is a cylindrical, vertical reactor vessel and is made up of three distinct sections. These sections are (from top to bottom): Stage 2, where the coal is injected; Stage 1, the position of the char burners; and the slag quench zone right at the bottom.

The walls of stage 2 are insulated to reduce heat loss to the environment. There are also vertical water tubes used to keep the temperature low enough to prevent hot spots and corrosion by H_2S.

Stage 1's outer walls are also refractory lined, but are bent inward to form a trough to assist in separating the molten ash from stage 1. The trough also serves to increase the gas velocity so that char or unreacted coal would not drop down from stage 2 to stage 1.

Coal is transported to the injector nozzles by recycled gas. A separate annulus in the nozzle conveys the steam, and the two streams only unite at the tip of the nozzle. This combined stream contacts the gases from stage 1 and gasification takes place.

Char is carried to the char burners via steam. They enter in stage 1 and the char is partially oxidized to render the gas which in turn moves to stage 2 where the incoming coal is gasified and subsequently entrained. The char burners are positioned to fire cyclonically, resulting in a swirling motion to increase gasification as well as separation of slag.

Excess steam is injected close to the trough to solidify entrained molten slag. This slag, together with that separated by the char burners, flows down the walls of stage 1 and is collected in the slag quench zone below.

The quench zone is filled with circulating water which shatters the molten slag to a granular form as it falls down from stage 1. The granular slag gravitates downward, where it is forced to the outlet nozzles through agitation by high pressure water jets. From here the slag is removed to slag lock hoppers for disposal.

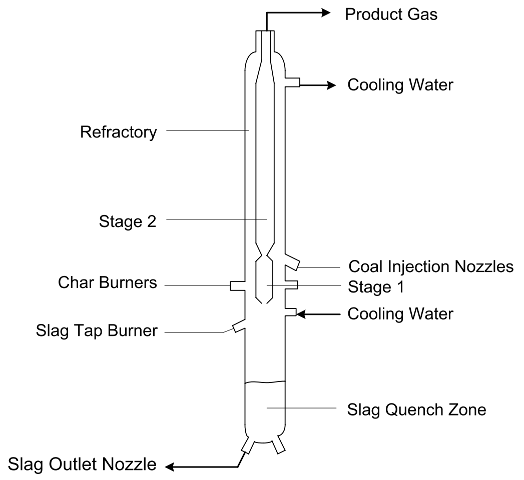


Figure 3.12: BIGAS Gasifier (adapted from Hartman et al. (1978))

Operating Conditions

Temperature:

- Stage 1: 1250°C to 1650°C
- Stage 2: 925°C
- Effluent gas: 815°C to 925°C

Pressure: 3.5 MPa to 10.4 MPa

Residence time:

- Stage 1: 2 seconds
- Stage 2: 8 seconds to 10 seconds

Reactants

Coal: Accepts most coal types, pulverised to 70 %, 200 mesh. Moisture content is not critical because the coal is slurried and pumped through the high pressure, slurry feed system.

Oxidant: Oxygen required at 0.5 ton oxygen per ton of coal.

Steam: Steam required at 0.4 ton steam per ton of coal.

3.3.3 Combustion Engineering Process

Project Development

Combustion Engineering instigates a process to produce an economical low energy content coal gas for electricity generation in an environmentally friendly method.

Two gasification systems were investigated – one operating at atmospheric pressure, the other at 10 atm. The gasifier at 10 atm was found to be as much as 8 % cheaper than the atmospheric one, mainly due to the reduced gasifier volume required. Conversely, the higher pressure gasifier will be more complex, requiring supplementary developments. Due to the number and extent of additions, the atmospheric gasifier was preferred.

Air was selected as original oxidant, with oxygen set to replace it at a later stage.

1974: Design of the process unit at Windsor, Connecticut commenced.

1977: Construction of the Windsor unit concluded.

Technology

The gasifier developed by Combustion Engineering Inc. is a cylindrical, vertical vessel operating at atmospheric pressure (figure 3.13). Coal enters the gasifier in pulverised form through nozzles situated at the bottom of the gasifier, known as the combustion zone. It is subsequently oxidized in this combustion section with a close to stoichiometrical quantity of air.

The combustor nozzles are arranged tangentially around the combustion section. The reduction section follows the combustion section to the top, where additional coal can be fed. Hot gases from the combustion section gasify and entrain the coal from the reduction section as it is transported vertically through the gasifier. Molten slag from the combustion zone is removed at the bottom and steam is generated in the refractory-lined walls covering the gasifier.

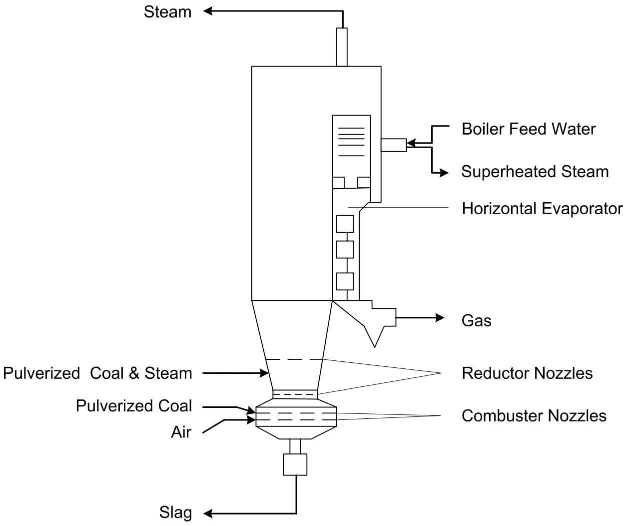


Figure 3.13: Combustion Engineering Gasifier (adapted from Hartman et al. (1978))

Operating Conditions

Temperature:

- Combustion zone: 1650°C to 1750°C
- Effluent gas: 925°C to 980°C

Pressure: atmospheric

Reactants

Coal: All types of coal, pulverised to 70 %, 200 mesh and dried to a moisture content of between 1 % and 2 % accepted.

Oxidant: Air required at about 4.5 ton of air per ton of coal.

3.3.4 Koppers-Totzek Process

Project Development

1948: The first pilot unit for gasifying coal in suspension is designed and erected by Koppers Company in conjunction with Heinrich Koppers. The gasifier was blown with oxygen and employed the Fischer-Tropsch process to produce synthesis gas.

1952: The Koppers-Totzek (K-T) gasifier goes commercial as the first two-headed unit is built in Finland.

1961: A gasification process is erected in Spain which tests the acceptability of feeding various types of coals to the gasifier. Fluid as well as delayed-type petroleum cokes were found to be gasified successfully.

1962: Most synthetic ammonia plants worldwide, using coal as feed, equipped with K-T gasifiers.

1969: The first four-headed gasifier (with approximately double the capacity of the two-headed gasifier) is installed in New Delhi, India.

Technology

The Koppers-Totzek gasifier is an ellipsoidal, horizontal reaction vessel. In the case of the two-headed gasifier, the heads are situated at the furthest ends of the ellipsoid, as in figure 3.14. The four-headed gasifiers consist of intersecting ellipsoids, with the burners placed at 90° angles.

The coal is transported from stockpiles, pulverised to between 70 % and 90 %, 200 mesh and dried to a range of 2 % to 8 % moisture. A variable speed screw feeder transports the coal to the gasifier, where it is pre-mixed with oxygen and steam before being injected into the gasifier.

The coal, oxygen, steam mixture enters the gasifier through flanking burners inside each gasifier head. To avoid flashback, the injection speed is maintained higher than that of the flame propagation speed.

The outer gasifier shell is refractory lined and jacketed. It contains water which absorbs the heat escaping from the gasifier and subsequently produces the steam required for gasification.

The coal is oxidized within a second of entering the gasifier. Ash contained in the coal is liquefied due to the reaction temperature being higher than that of the ash fusion temperature. Approximately half of the ash falls down into the slag quench tank as molten slag. The quench is filled with circulating water which shatters the molten slag to a granular form. The rest of the ash is entrained with the gas as fine slag particles, where they are solidified by water sprays at the gasifier exit.

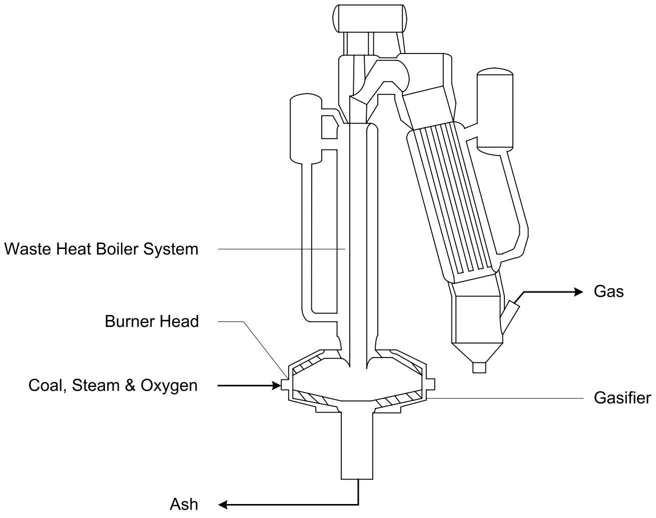


Figure 3.14: Koppers-Totzek Gasifier (adapted from Hartman et al. (1978))

Operating Conditions

Temperature:

- Combustion zone: 1925℃
- Effluent gas: 1500℃

Pressure: 3.5 kPa or 35 kPa to 50 kPa

Residence Time: Coal gasified in under a second.

Reactants

Coal: All types of coal are accepted regardless of rank. Coal is pulverised to between 70 % and 90 %, 200 mesh and dried to a moisture content of between 2 % and 8 %.

Oxidant: Oxygen required at between 0.85 and 0.9 ton per ton of bituminous coal and between 0.7 and 0.75 ton per ton of ligite.

Steam: Steam required at between 0.3 and 0.34 ton of steam per ton of coal.

3.4 Molten Bath Gasifiers

In molten bath gasifiers, the feed coal is crushed, mixed with steam and oxygen or air and injected into a hot liquid bath. The fixed carbon contained in the coal dissolves in the hot bath, reacts with the oxygen and is subsequently removed as carbon monoxide. The unreacted carbon floats on the molten bath surface along with ash, from where it is ejected. Molten salt, coal ash or iron are the typical compounds used in molten bath gasifiers.

Two molten bath gasifiers will be discussed.

3.4.1 Rockwell International

Project Development

1974: Atomics International, a division of Rockwell International Corporation performs experiments to develop molten salt combustion and gasification processes. They found that the processes were acceptable and that a pilot plant should be built to test suitability of commercialization.

1975: Preliminary designs show that project costs would be higher than anticipated. To lower the total costs involved, it was decided not to erect the plant at the original location of Norwalk Harbour, Connecticut, but to rather build it in Santa Susana, California.

1976: Atomics International commences the development of a molten salt coal gasification process to illustrate the feasibility of this technology to generate electricity in an environmentally friendly manner. Research included testing the suitability of molten sodium carbonate at pressures as high as 3 atmospheres and temperatures of 1150℃.

1978: Initial testing conducted with petroleum coke, at near atmospheric conditions. No melt was regenerated. Successive tests suggested that the electric heaters for melt removal should be replaced by gas burners. These were subsequently designed and installed.

Technology

The molten bath (salt) gasifier is of cylindrical, vertical orientation (figure 3.15). The walls of the gasifier are lined with α-alumina refractory bricks to prevent corrosion from the molten salt, while heat loss is reduced by an insulating layer between the gasifier wall and refractory bricks.

The gasifier is fitted with four downward facing nozzles, used to inject the air-solids reagents as well as a horizontal melt removal compartment.

The feed to the gasifier consists of coal and sodium carbonate, transported by air into the molten salt bath. The coal partly oxidises and pulverises upon entering the hot melt. The product gas escapes while the melt retains the sulfur and ash contained in the coal. This has as advantage that the product gas does not have to be cleaned of the ash and sulfur before use.

The main functions of the sodium carbonate are as follows:

- Serves as dispersing instrument for the coal and air to be gasified.
- Serves as heat sink, and heat source for the pyrolysis of the coal.
- Absorbs the sulfur contained in the coal and retains the ash.
- Serves as location where the sulfur compounds can act as catalyst for the oxidation of coal.

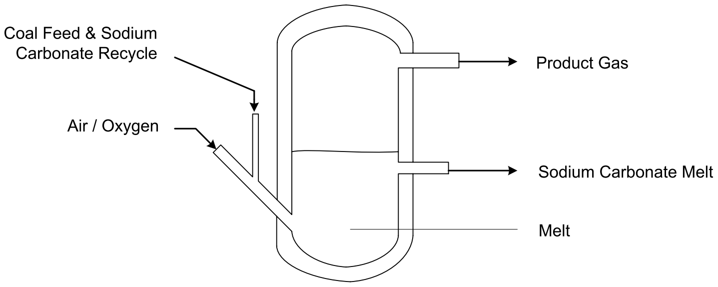


Figure 3.15: Rockwell International gasifier (adapted from Hartman et al. (1978))

Operating Conditions

Temperature:

- Melt bed: 980℃
- Effluent gas: 980℃

Pressure: 100 kPa to 1930 kPa

Superficial gas velocity: 0.5 m/s to 0.7 m/s

Reactants

Coal: All types of coal are accepted. There is no requirement of pre-treatments such as sizing, fines removal or pulverization. Crushing is suggested to enable pneumatic transport. Moisture content should be less than 10 % to reduce plugging of solids.

Oxidant: Successful gasification requires between 3.5 and 4.0 tons of air per ton of coal, or between 0.66 and 0.70 tons of oxygen per ton of coal.

Steam: No steam is required for air blown gasification, while oxygen blown gasification requires between 0.1 and 0.4 ton of steam per ton of coal.

3.4.2 Saarberg/Otto

Project Development

1950: Union Kraftstoff constructs the first Rummel/Otto gasifier in Wesseling, Germany.

1956: Union Kraftstoff installs a single shaft gasifier, with an output capacity more than 4 times that of the original gasifier. Apart from the increased capacity, the gasifier is oxygen blown, runs on brown coal and can process liquid fuels.

1950s: Experimental work is conducted on the development of a double-shaft gasifier. This gasifier uses the room between the two concentric shafts for gasification and combustion. This annular space is divided into two compartments, with a molten slag bath flowing between these compartments. The gasifier performance was however disappointing, because more than 60 % of the product gas was made up of nitrogen. Attempts to rectify this only resulted in the rate of gas production dropping considerably.

1960: Dr. C. Otto and Company, who by this time had sole responsibility for the Wesseling plant, designed an advanced edition of the single shaft gasifier. Saarberg/Otto also illustrated operation at high pressures (2500 kPa), as well as the use of varying types of feedstock and the production of diverse product gases.

1962: Despite the under-performance of the double-shafted gasifier, such a gasifier was constructed in Bromley, England. The gasifier ran on Midland coal, but the operation was suspended following design and operational difficulties.

1964: Steam/naphta reformers were installed to reduce the cost of the synthesis gas, causing the whole coal gasification plant to be shut down.

Technology

The Saarberg/Otto, single-shaft gasifier is a cylindrical, vertical vessel made up of three distinct sections – see figure 3.16. The first section is situated at the bottom and is a molten slag whirlpool. Section 2 follows on that and is the reaction zone, with the third section, gas cooling right at the top. A tubular wall surrounds sections 1 and 2, mainly to reduce heat loss.

The first section is fitted with side nozzles to inject coal, ash, char and the oxygen and steam required. The coal undergoes drying and grinding before being housed in a charging hopper, from where it is pneumatically fed to the injection nozzles via recycled gas. The nozzles are fitted in a downward sloping direction, toward the molten slag bath's surface. The slag bath rotates rather slowly due to the momentum of the coal, steam and oxygen mixture, which enhances the reaction rates. The slag bath further serves as heat shield and oxygen transfer medium.

All of the finer coal particles are gasified in section 1. The resulting gas spirals upward through a trough in the second section, while some of the entrained slag and char particles are removed by the centrifugal action and falls back into the slag bath. The remainder of the entrained char is gasified additionally in section 2.

The product gas moves further upward to section 3 where the liquid slag droplets are solidified upon contacting the recycled gas, where after the gas exits the gasifier. Char and slag particles contained in the exit gas are recovered in a cyclone and subsequently recycled to the gasifier.

The slag gathered in the first section is transferred through a tap in the base of the gasifier, quenched, shattered to granular form in the water bath and sent for removal.

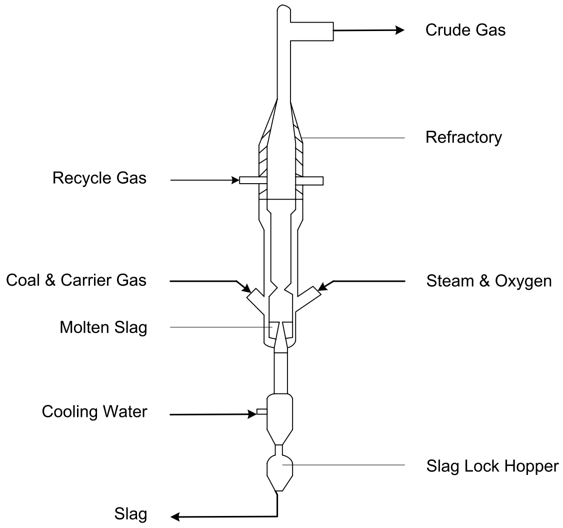


Figure 3.16: Saarberg/Otto gasifier (adapted from Hartman et al. (1978))

Operating Conditions

Temperature:

- Gasifier: 1500℃ to 1700℃
- Effluent gas: 815℃ to 925℃

Pressure: 2500 kPa

Gas velocity: 1.8 m/s above the slag bath

Reactants

Coal: All types of coal, pulverized to 100 %, 16 mesh are accepted. Moisture content is not critical.

Oxidant: Successful gasification requires approximately 1 ton of oxygen per ton of coal.

Steam: Successful gasification requires approximately 0.4 ton steam per ton of coal.

The location of some of the gasifiers just mentioned is shown on the coal deposition map in figure 3.17. Most of these are situated in the western parts of the United States of America.

Figure 3.17: Gasifiers around the world

Chapter 4

Modelling

4.1 Definition

In the "Defense Acquisition University Press", a model is defined as: "a physical, mathematical, or logical representation of a system, entity, phenomenon, or process." When engineers study systems with the objective to optimize or control, they typically use mathematical models. Mathematical models enable engineers to construct clarifying models of systems as hypotheses of how these systems function, or they attempt to predict how an unanticipated occurrence may influence the system.

Eykhoff (1974) defined a mathematical model as: "a representation of the essential aspects of an existing system (or a system to be constructed) which presents knowledge of that system in usable form." Mathematical models typically use variable sets to portray the properties of systems, i.e. system outputs, counters, etc. These variables are linked via sets of equations giving the relationships among them. The function sets describing these relationships make up the final model. These models have various varieties, and systems are typically represented by dynamic, differential or even statistical techniques. The models are however not limited to any one of these generic types, but are often combinations of these.

4.2 Modelling procedure

There are many ways to approach modelling problems, while physical systems can also be represented by a number of different techniques. A logical, step wise procedure for performing such modelling exercises is provided by Marlin (2000). His method is based on the fundamental laws of conservation of mass, energy and momentum. Making use of these fundamental laws and theories has several distinct advantages:

- Very few principles are required to sufficiently describe a wide range of physical systems.
- Our view of nature is simplified.
- Fundamental models have a broad range of applicability – extrapolation beyond the regions of immediate empirical experience is therefore possible.
- They provide analytical expressions, relating important features of the physical system to its dynamic behaviour.

The procedure prescribed by Marlin (2000) follows:

1. Define goals
   (a) Specific design decisions
   (b) Numerical values
   (c) Functional relationships
   (d) Required accuracy
2. Prepare information
   (a) Sketch process and identify system
   (b) Identify variables of interest
   (c) State assumptions and data
3. Formulate model
   (a) Conservation balances
   (b) Constitutive equations
   (c) Rationalize (combine equations and collect terms)
   (d) Check degrees of freedom
   (e) Dimensionless form
4. Determine solution
   (a) Analytical

   (b) Numerical

5. Analyze results
   (a) Check results for correctness
      i. Limiting and approximate answers
      ii. Accuracy of numerical method
   (b) Interpret results
      i. Plot solution
      ii. Characteristic behaviour like oscillations or extrema
      iii. Relate results to data and assumptions
      iv. Evaluate sensitivity
      v. Answer "what if" questions
6. Validate model
   (a) Select key values for validation
   (b) Compare with experimental results
   (c) Compare with results from more complex model

These steps do not have to be followed rigorously, but should be seen as guidelines to assist someone in modelling a process successfully. The steps can be divided into two groups: steps 1 to 3 – model development and steps 4 to 6 – simulation and model solution. Although the steps are shown in order, it is often the case that the modeller has to iterate to arrive at a satisfactory solution. This is why experience is vital to model systems well.

4.2.1 Define Goals

One of the vital steps in process modelling is the decision regarding the type of model required. Goals should be as specific as possible regarding the information required. A specific numerical value may be desired, for example "At what time will the tank overflow?" Apart from explicit numerical data, semi-quantitative information about the system's characteristics and/or behaviour may be required, for example "Will the level increase monotonically or will it be oscillatory?" Additional insight regarding functional relationships will also be necessary, "How will the flow rate impact the time that the overflow will occur?"

The required model accuracy is another aspect that should be considered when setting goals, including the effects these inaccuracies will have on the obtained results. Inaccuracies are dealt with by identifying likely errors in assumptions and data at the onset and then observing their effects throughout the rest of the modelling and finally the various analyses.

4.2.2 Prepare Information

Proper identification of the system is the first task at hand. This is done by sketching the system, defining the system boundaries and identifying the key variables for which the necessary balances should be formulated. Partial differential equations are commonly employed to model processes in which important variable values vary with position. Systems without any spatial fluctuations regarding variable values are deemed lumped-parameter systems, while systems exhibiting significant deviations in spatial directions termed distributed-parameter systems.

Apart from the initial system selection, models also require additional information in order to predict the system's behaviour. The assumptions made to base the model on are some of the important considerations to be kept in mind when considering the information required. Thorough investigation of the physical system and the subsequent level of accuracy required to satisfy the goals set in part 1 are considered in order to make the assumptions. Engineers are typically not bothered by system descriptions on atomic or microscopic level, but rather interested in their macroscopic behaviour. The macroscopic description is also more often than not quite adequate to comprehend the process dynamics. The assumptions made usually trade off modelling accuracy (preferring detailed models) with solution, which normally favours simpler model descriptions.

The next element of information gathering is the physical and chemical data describing the system, such as reaction rates, heat capacities and densities. The external inputs (forcing function) should also be defined, so that changes in the system's behaviour can be tracked successfully (such as fouling of a heat exchanger).

4.2.3 Formulate the Model

First of all, the variables deemed important and whose behaviour should be tracked and/or predicted are to be selected. The appropriate equations are then developed from fundamental principles. These can be classified as either conservation equations or constitutive equations. Conservation equations describe the relationships among the physical properties of the selected system. The conservation of material, energy and momentum are the most important of the conservation equations. Their general form is that of equation 4.1:

$$\text{Accumulation} = \text{In} - \text{Out} + \text{Generation} - \text{Consumption} \tag{4.1}$$

For cases where the accumulation term is zero, equation 4.1 reduces to an algebraic equation, while for nonzero accumulation, the equations are ordinary differential equations. The equations are selected to convey the maximum amount of information on the selected dependent variables. Selection of the correct balances can be facilitated using the following guidelines:

- If the variable in question is a total mass or pressure, a material balance should be used.
- If the variable in question is a concentration or weight fraction of a component, the component material balance is required.
- If the variable in question is a temperature, the appropriate energy balance should be derived.

The model may however also be developed to track and/or predict the behaviour of multiple variables. In these situations it is often the case that there is an insufficient number of balances to describe all the variables. This is when additional constitutive equations are required and included in order to render the number of equations sufficient and the model completely specified. These constitutive equations are relationships that are not universally valid, but rather chosen to be only sufficiently accurate as for the system required.

The number of equations required to describe the system can be obtained from a degrees of freedom analysis. A complete system description should have no degrees of freedom (DOF), as formulated in equation 4.2:

$$\text{DOF} = \text{NV} - \text{NE} \tag{4.2}$$

with NV the Number of dependent Variables, and NE the Number of independent Equations.

4.2.4 Mathematical Solution

Various possibilities exist to solve a mathematical problem. The technique to use should be the one that provides greatest insight into the system. Analytical techniques are consequently often used. The main reasons are:

- They can calculate specific numerical values.
- They determine the important functional relationships among the different variables as well as the system behaviour.
- Insight is gained into the sensitivity of the results to changes in the data.

These advantages are often so vital that certain simplifying assumptions are made to enable the use of analytical solutions, such as linearization of the non-linear terms. These assumptions are however not always valid and may in some cases yield unacceptable errors. When this occurs, numerical solutions are used. Numerical solutions are unfortunately not exact, but the introduced errors can generally be made small enough to give better results than with the assumptions for analytical solutions. The disadvantage of numerical solutions is therefore mainly the loss of insight into the system.

4.2.5 Results Analysis

Results analysis aims to ascertain whether the obtained solution is indeed correct. This is verified by checking if the solution conforms to certain criteria, such as:

- Satisfying initial as well as final conditions.
- Stays within certain bounds.
- Has negligible numerical errors.
- Conforms to semi-quantitative expectations, i.e. signs of output changes.

Extensive analysis and interpretation of the results are also important. Key features should be related explicitly to certain parameters to sufficiently understand their behaviour. Sensitivity in the results to changes in the data and/or assumptions should also be validated (what if analysis).

4.2.6 Validation

The validation step entails the verification of steps 1 to 5 to ascertain how well they represent the physical process within the required range of conditions. Even though the analysis of step 5 should provide the necessary certainty that the results are in fact correct, comparisons with empirical data are required in order to adequately validate the model. Good comparisons are however not sufficient to render a model adequate, but rather demonstrate that the data does not invalidate the model. An infinite number of experiments would be required to cover the full spectrum of possible conditions and no model can therefore be validated completely. The system can however be characterised sufficiently for a specified range of operating conditions.

4.3 Comparing gasifier features

Table 4.3 was compiled from the gasifier information covered in chapter 3. It provides a detailed layout of the various characteristics associated with these gasifiers.

The reagents column in table 4.3 is interpreted as follows: The two vertical lines separate the gasifier into three regions – top, side and bottom sequentially. The letters are then used to indicate where each reagent enters the gasifier, i.e. "c ⋮ s o ⋮ s o" represents coal entering at the top, with steam and oxygen being able to enter at either the side or the bottom.

4.3.1 Geometry

All but one of the gasifiers, the Koppers-Totzek gasifier, are of cylindrical and vertical orientation, with the Koppers-Totzek gasifier being ellipsoidal and horisontal. Fixed bed gasifiers operate using the packed tubular bed principle. These gasifiers are typically longitudinal to prolong the contact between the different reacting phases. The extent of reaction and conversion of reagents are strongly dependent on the length of the reactor, as illustrated in figure 4.1 which shows the relationship between the reaction rate and conversion. The area underneath the graph denotes the reactor volume required to achieve said conversion. The Koppers-Totzek gasifier is of the entrained flow type, and as such contact between the reagents occur in a uniform manner, which is similar to a continuously stirrer tank reactor (CSTR). The length of these gasifiers does not have the same effect on conversion as the tubular bed's and is therefore not as important.

Table 4.1: Summary of gasifier characteristics

Industrial Gasifiers	Section	Gasifier bed	Subsections	Water Jacket	Pressure (MPa)	Temperature range (°C)	Reagents		
							Top	Side	Bottom
British Gas/Lurgi Slagging	3.1.1	Fixed	4	yes	0.5-2.6	400-1300	c	s o	
Lurgi, Dry Ash	3.1.2	Fixed	3	yes	2.7	720-1150	c		s o/a
GFERC Slagging	3.1.3	Fixed	3	no	n/s	n/s	c		s o
Wellman-Galusha	3.1.4	Fixed	2	yes	0.1	620-1315	c		s a
Woodall-Duckham	3.1.5	Fixed	2	yes	0.1	120-1200	c		s a
Battelle Ash Agglomerating	3.2.1	Fluidized	3	no	0.7-0.9	870-890	c a	s	s
Carbon Dioxide Acceptor	3.2.2	Fluidized	2	yes	1.0	n/s	a		c s
Hygas	3.2.3	Fluidized	4	no	7.9-8.1	315-1000	c		s o
Synthane	3.2.4	Fluidized	2	n/s	4.1-6.9	815-980	c	s o	s o
Winkler	3.2.5	Fluidized	2	no	0.1-0.4	980-1150		s o/a	c s o
Babcock and Wilcox	3.3.1	Entrained	1	no	0.1-2.0	980-1870			c s o
BI-Gas	3.3.2	Entrained	3	yes	3.4-10.4	925-1650		c s o	
Combustion Engineering	3.3.3	Entrained	3	yes	0.1	1650-1750			c s a
Koppers-Totzek	3.3.4	Entrained	1	yes	0.04-0.05	1500-1925		c s o	
Rockwell International	3.4.1	Molten bath	2	no	0.1-1.9	980		c s o/a	
Saarberg/Otto	3.4.2	Molten bath	3	n/s	2.5	1500-1700		c s o	

with c = coal; s = steam; o = oxygen; a = air; o/a = oxygen/air
n/s = not specified

Industrial Gasifiers	Flow	Coal	Situated	Coal residence time	Gas velocity (m/s)
British Gas/Lurgi Slagging	semi-batch	bituminous,caking	Westfield, Scotland	10-15 min	0.75
Lurgi, Dry Ash	semi-batch	almost any	Secunda, South Africa	1 h	
GFERC Slagging	semi-batch	noncaking, high moisture (35 %)	Grand Forks, North Dakota	-	-
Wellman-Galusha	continuous	anthracite, bituminous, coke, sub-bituminous	Reading, Pennsylvania	4 h	fluctuates
Woodall-Duckham	continuous	non or weakly caking	Milan, Italy	several hours	fluctuates
Battelle Ash Agglomerating	semi-batch	non-caking	West Jefferson, Ohio	n/a	n/a
Carbon Dioxide Acceptor	semi-batch	sub-bituminous, lignite	Rapid City, South Dakota	n/a	n/a
Hygas	continuous	any	Chicago, Illinois	n/a	n/a
Synthane	continuous	any	Bruceton, Pennsylvania	n/a	n/a
Winkler	continuous	lignite, sub-bituminous	Leuna, Germany	n/a	n/a
Babcock and Wilcox	continuous	most types, preferably dried	Morgantown, West Virginia	1s	4.5-9
BI-Gas	continuous	any	Homer City, PA	2-10 s	-
Combustion Engineering	continuous	any	Windsor, Connecticut	-	-
Koppers-Totzek	continuous	any	New Delhi, India	1 s	-
Rockwell International	semi-batch	any	Santa Susana, California	0.5-0.7	n/a
Saarberg/Otto	semi-batch	any	Wesseling, Germany	1.8	n/a

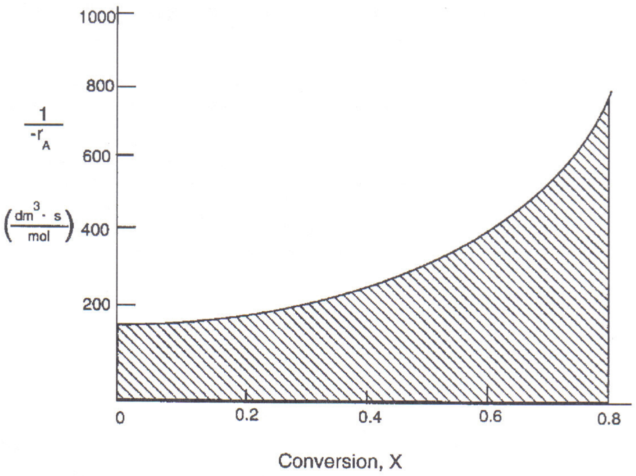


Figure 4.1: Size of tubular reactors versus conversion (Fogler, 2002)

The geometry of gasifiers impact modelling with regard to the type of equations required to model the reactions. The equations used to calculate the reaction rates achieved depend on whether the gasifiers emulate tubular beds or CSTRs.

The equation generally used to calculate the reaction rate achieved in tubular beds is:

$$\frac{dF_A}{dV} = r_A \tag{4.3}$$

and for CSTRs:

$$\frac{F_{A_0} - F_A}{V} = -r_A \tag{4.4}$$

4.3.2 Principal gasifier types

Through this investigation it became apparent that there are four main gasifier categories. They differ mainly regarding the mechanism used for bringing the reagents into contact with one another, i.e. the gasifier bed. As mentioned before, the main classification is as follows:

- Fixed bed gasifiers
- Fluidized bed gasifiers

- Entrained flow gasifiers
- Molten bath gasifiers

When investigating these generic types, certain differences are observed, these are summarised in table 4.2.

From table 4.2 it is seen that fixed bed gasifiers are not only the best developed, but also generally employ the most complex gasifier technology. Complexity considers both internal mechanics and pre-treatment of reagents. Conversely, the molten bath gasifiers are the least developed but also the least complex. Removal of the unwanted products (mainly ash) is a problem that should be kept in mind in all gasifier designs. The modelling of gasifiers should also take this important aspect into consideration. A variety of operating temperatures are associated with the different types, but discrepancies are present even among gasifiers of similar type. Classification based on temperature among the generic types is therefore trivial and will rather be discussed for specific gasifiers.

Process complexity and maturity is dealt with in modelling by keeping in mind that the more complex a process, the more detail is required to model it accurately and modelling inaccuracies are therefore rather probable. Maturity on the other hand provides the modeller with numerous well developed systems which can be studied and used to compare data with. Validity of the data of these systems are therefore also considered to be high.

4.3.3 Operating conditions

Figure 4.2 is derived from table 4.3 and shows the normal operating regions (temperature and pressure) of the studied gasifiers. It is seen that there are several regions of mutual operation, but also regions where only specific gasifiers operate. Several gasifiers have a large operating range regarding either/or temperature and pressure, while certain others only operate at specific or small ranges of pressures. The gasifiers are seen to generally have larger ranges associated with operating temperature, than they do regarding pressure. The majority of gasifiers operate between atmospheric pressure (101.325 kPa) and about 10 000 kPa, while the temperatures range between 120℃ and 1900℃.

An important modelling consideration is the fluid package to be used. This choice is generally influenced by the desired operating temperature and pressure. By using

Table 4.2: Comparing gasifier features of main gasifier types

Attribute	Fixed bed	Fluidized bed	Entrained bed	Molten bed
Maturity	Well established with many commercial designs	Less developed than fixed bed	Less developed than fixed bed	Least developed of all types
Complexity	High level of mechanical complexity (internal moving parts)	No internal moving parts. Design of distributor plate critical	No internal moving parts. Geometry less complicated than fluidized bed	Normally less complex, although bath circulation increases complexity
Treatment	Requires sized coal, fines handled separately	Fines should be removed to prevent elutriation	No fines declined	No pre-treatment required
Product gas	Multi-stage cleanup required	Superb gas-solid contact	Sensible heat in gas has to be recovered – decreases efficiency	Entrainment of alkali particles can prevent usage of synthesis reactors
Ash disposal	Ash contains small amounts of carbon	Dry ash gravitates to bottom – contains more carbon	Increased thermal loss in ash	Ash contains fairly high carbon quantity and should be separated from catalyst which is recycled
Temperature	Countercurrent flow causes temperature gradient. Low thermal losses	Can operate at lower temperatures	Highest temperatures off all categories	Temperatures achieved can be corrosive to refractory

figure 4.2 to ascertain the operating conditions of a gasifier to be modelled, an informed choice can be made regarding the most appropriate fluid package to be used.

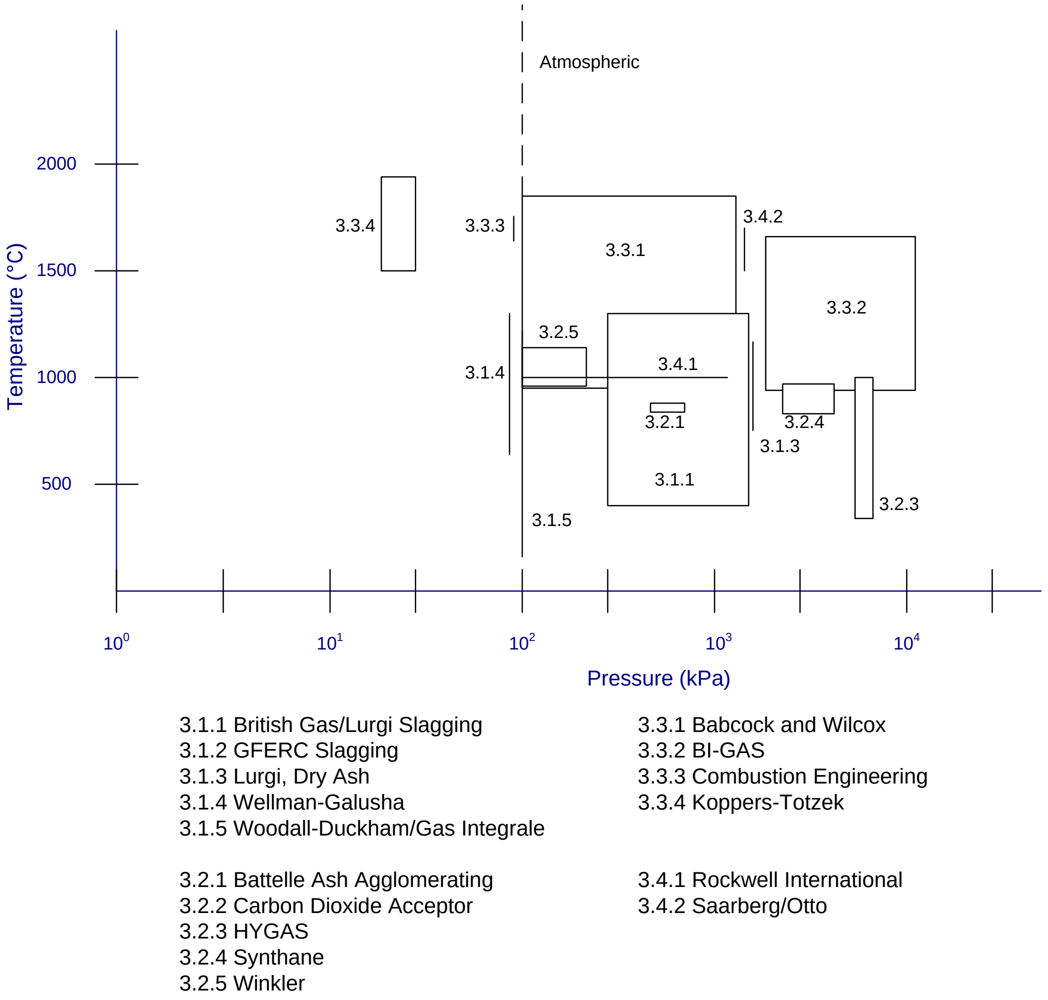


Figure 4.2: Pressure-Temperature operating regions of various gasifiers

4.3.4 Mode of operation (Continuous/Batch)

Continuous processes operate constantly, whereas batch processes are intermittent. The mode of operation of a process has very important consequences on the modelling exercise. The flow of material and the building blocks used to represent the process are all dependent on whether the processes are run in batch or continuous mode. Most processes involving the processing of solids have some sort of batch character. In gasification this is no different. None of the gasification processes investigated operate on an exclusively batch basis, but rather a combination of the two, namely semi-batch. The feed of the coal into certain gasifiers happen on a semi-batch basis. Because the gasifier has to be maintained at a constant pressure, the coal is inserted via hydraulic valves that pressurise the coal housing chamber prior to loading into the gasifier. The remainder of the processes, including the removal of the effluent all occur continuously.

Gasifiers where the coal is inserted in a pulverised form and injected together with air and/or oxygen streams can however operate on a continuous basis.

4.3.5 Oxidant

The type of oxidant used is another factor which influences the modelling of gasifiers immensely. Oxygen is the agent required for gasification. It is however not always injected in its pure form, but regularly enters the gasifier as air. Pure oxygen gasification produces a higher energy content effluent, which is desired in power generation operations. The effluent is also free from any nitrogen based compounds.

For modelling purposes, it should be kept in mind that pure oxygen gasification produces more energy than similar air blown gasification, while the compounds associated with air blown gasifiers complicate the modelling by providing compounds whose flow rates should be calculated additionally.

4.3.6 Points of entry

The gaseous reagents, steam and oxygen or air generally enter the gasifiers at either the bottom or on the side. These then diffuse upward to contact the coal which mostly enters from the top and descends downward via gravitation. In some cases, the coal is pulverised prior to entry and is then injected into the gasifier together with some of the reagent gases. In these cases, the point of entry is mostly on the side or bottom of the gasifier.

The relevance of the point of entry for modelling purposes is the direction of contact between the different phases, i.e. co-currently or counter-currently. These two methods of contact have differing effects on the temperatures attained as well as residence times required to attain the desired conversions. The sequencing of the building blocks used to represent the system is also affected whether the contact occurs co-currently or counter-currently. See figure 4.3 for an example of a counter-current gasifier representation.

The phases (liquid, gas, solid) of the compounds entering the gasifier, should be specified in the modelling environment, as different entering phases will yield differing

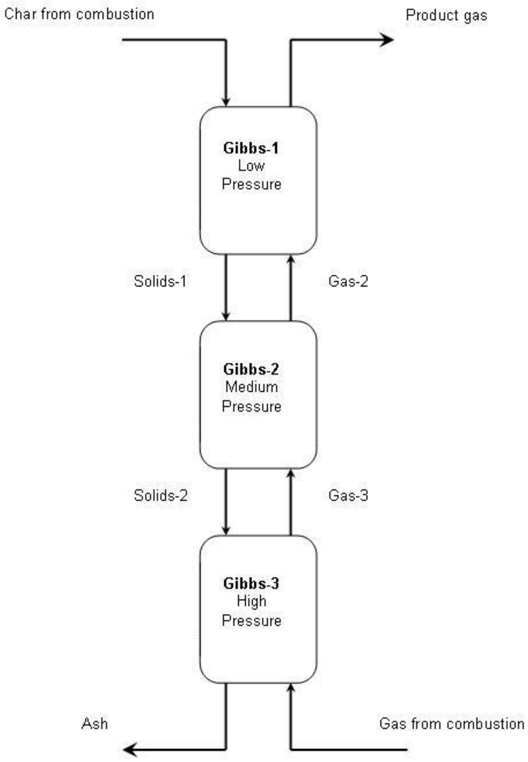


Figure 4.3: Modelling representation of a counter-current gasifier (Clack, 2008)

results.

4.3.7 Type of coal

Because coal does not have a unique composition, it does not exist as a single entity in modelling environments. The coal composition is generally used and the components involved entered in elemental form. For accurate modelling, the free swelling index, water content as well as any other unique characteristics of the coal used should be taken into account.

4.3.8 Number of internal subsections

For each distinct section in a gasifier, a different modelling approach may be required. The gasifier is divided into these sections and each one then modelled/simulated successively. When combined, they should ultimately model the gasifier sufficiently.

4.4 Kinetic Considerations

The reactions and kinetics associated with gasifiers are another crucial aspect to be investigated for successful modelling. Various researchers have performed experiments, studying gasifier kinetics. Their contributions are summarised here.

4.4.1 Research on gasifier kinetics

Denn and Yu

In their research, Denn & Yu (1979) performed detailed kinetic experiments on moving bed coal gasifiers. Various gasification experiments were conducted with the principal aim to establish the relevance of the coal particle size. They focused on using Wyoming coal with two different sized particles (3 mm and 100 mm). The conditions of gasification are however unknown. Their experiments revealed that the combustion front is significantly widened by the mass transfer limitation, which consequently reduces the maximum obtained temperature by approximately 150℃. The effluent gas temperature and composition however remained virtually unaffected. The general performance is subsequently seen to be nearly constant despite changes in the physico-chemical processes, and therefore deemed to be insensitive to changes in the reactions. Denn & Yu (1979) therefore suggested that a “kinetics-free” model could be used to estimate gasifier effluent characteristics.

Their kinetics-free model compared well with a more detailed model proposed by Yoon et al. (1976) (table 4.3). The models agree especially well in regions of high carbon conversion and for good coals with negligible reactivity towards hydrogen. It should be noted that Denn & Yu (1979) did not consider the formation of methane.

Table 4.3: Comparison of effluent concentrations (moles) (Yoon et al., 1976)

Component	Yoon	Denn
H_2O	0.428	0.406
H_2	0.525	0.594
CO	0.332	0.356
CO_2	0.328	0.328
CH_4	0.025	0
T (℃)	762	734

In regions of the maximum gasifier temperature, the oxidation reactions generally become mass transfer limited. The particle size therefore affects the model prediction significantly in these regions. It was also found that the exit gas composition and temperature is also fairly insensitive to oxidation selectivity. The performance of the gasifier in regions of effective carbon utilization is therefore only dependent on thermodynamic constraints and stoichiometry.

The composition and temperature of the effluent gas can be predicted without detailed kinetics, by making only three simplifying assumptions:

- Carbon conversion is known.
- The water-gas shift reaction is at equilibrium.
- Complete conversion of oxygen in feed.

With the kinetics-free model proposed, it is possible to make fast predictions of the overall gasifier performance, specifically regarding the effluent gas composition and temperature. Optimum results are found for coals with negligible hydrogen reactivity and when sufficient utilization of carbon occurs.

Kosky and Floess

Kosky & Floess (1980), Yoon et al. (1978), Yoon et al. (1979a), Arri & Amundson (1978) found that coal gasifiers can be modeled adequately by applying detailed information of the local kinetics and chemistry. Although it is difficult to specify exactly which reactions take place inside a gasifier, these researchers have reached agreement on the primary reactions:

$$\begin{aligned} C + H_2O &\rightleftharpoons CO + H_2 && (4.5)\\ C + CO_2 &\rightleftharpoons 2CO && (4.6)\\ C + 2H_2 &\rightleftharpoons CH_4 && (4.7)\\ CO + H_2O &\rightleftharpoons CO_2 + H_2 && (4.8) \end{aligned}$$

These reactions are assumed to occur in the gasification zone, while oxidation is thought to take place directly below the gasification zone:

$$\begin{aligned} C + \frac{1}{2}O_2 &\rightleftharpoons CO && (4.9)\\ C + O_2 &\rightleftharpoons CO_2 && (4.10) \end{aligned}$$

Kosky & Floess (1980) state that the feed rate of coal into a gasifier is predominantly dependent on the reaction rate of the coal with the reacting gases. Internal adjustments are consequently made according to consumption.

Table 4.4: Comparison of effluent concentrations (Dry Volume %)

Component	Yoon	Kosky (H_2 in char)
CO	21.5	20.4
CO_2	26.8	25.1
H_2	42.5	47.0
CH_4	8.0	5.8
(others)	1.2	1.7

Table 4.4 compares the predicted effluent composition of the model developed by Kosky & Floess (1980) with the more complicated model derived by Yoon et al. (1978). The two models are seen to compare well. The advantages of the Kosky and Floess model are its simplicity and insensitivity to the kinetic parameters. Accurate prediction of the effluent gas temperature was however not possible.

The Kosky and Floess model calculates the output conditions of a fixed bed gasifier accurately, when using either air or oxygen. The assumed exit shift equilibrium is seen to be sufficient, considering the large number of possible gasification reactions. The prediction of methane production is however not satisfactory.

Britten

Britten (1988) showed that overall carbon conversion and gas production is established only by the conditions that cause the endothermic gasification reactions to be extinguished. The research focused on counter-current, moving-bed, char gasifiers.

An analytical model for the conversion in the gasifier (in terms of the asymptotic extinction temperature of the gasification reactions) was devised by lumping the gasification agents CO_2 and H_2O together with the products CO and H_2 into pseudospecies. Assuming that the water-gas shift equation reaches equilibrium at this temperature enables the prediction of detailed effluent gas composition leaving the reaction zone. The model was found to be valid for chars with adequately high reactivity.

Gas-solid reactions taking place inside gasifiers are usually far away from equilibrium, which suggests that the reactions are rate limited (Britten, 1988). Britten tried to determine the controlling factors influencing carbon conversion, and to develop a model that will predict the optimum char feed rate as well as effluent gas flow rate and composition. He found that, in general, overall gasifier conversion is only dependent on the kinetic extinction of the highly active endothermic gasification reactions.

Britten employed a counter-current, one-dimensional, adiabatic, packed-bed gasifier in his model. The gas (consisting of oxygen, steam and nitrogen) flows upwards, contacting the downward settling char (carbon and ash).

Britten's model is compared to that of Yoon et al. (1979a) in table 4.5 (using Wyoming coal). The predicted effluent composition and temperature compare favorably with the more comprehensive numerical model of Yoon et al. (1979a). Britten claims that the sources of the discrepancies can be identified and negated.

Table 4.5: Comparison of effluent concentrations (moles)

Component	Yoon	Britten
H_2O	0.334	0.381
CO_2	0.203	0.191
H_2	0.312	0.302
CO	0.150	0.125
T (K)	1023	1027

Chen

Chen et al. (2000) developed a detailed entrained flow gasifier model. In their studies they showed that recycle of the char is necessary to obtain 100 % carbon conversion for an air blown gasifier.

For the modelling of the gas-solid reactions and flow, the following chemical and physical phenomena were considered:

- Turbulent gas flow.
- Entrainment of particles (with turbulent dispersion).
- Devolatilization of coal and combustion of volatiles.
- Heterogeneous char reactions (combustion as well as gasification).
- Convective and radiative heating and/or cooling.

The coal is assumed to be made up of char, volatiles and ash. Chen et al. further assumes that the gaseous reactions are limited by the rate of reactant mixing and not by reaction kinetics, thereby enabling the calculation of the gas composition from local, immediate equilibrium by minimising the Gibbs free energy (as a function of local elemental compositions and energy values).

Additional assumptions were:

- Uniform inlet velocity and temperature distributions.
- Fully developed effluent flow conditions.
- No slip wall conditions.

The reactions occurring in the entrained gasifier are partitioned into three zones:

- Combustion (below the char burner).
- Gasification (between char burner and reductor burner).
- Devolatilization (above reductor burner).

Chen et al. found that the characteristics of these three regions control the performance of the gasifier.

The impact of the water-gas shift reaction was investigated by omitting it from the model calculations. The oxygen concentration did not differ from the previously calculated value, while the gas effluent temperature and concentrations of the other components revealed only small variations. Chen et al. state that additional research is required to quantify the interactions among the water-gas shift reaction and the other turbulent fluctuations.

Norman

Norman et al. (1997) developed a model to predict the environmentally unfriendly components (sulfur, nitrogen, chlorine and other trace elements) formed during gasification. Experiments focused on two generic gasifiers – either oxygen blown or air blown, and were tested on a variety of conditions. The model proved accurate for temperatures up to 1100℃ with accuracy decreasing for higher temperatures.

The composition of the gasifier effluent is dependent on various factors:

- The type of coal fed to the gasifier.

- The temperature profile in the gasifier.
- The gasifier pressure.
- The design of the process.
- Control devices downstream (i.e. pollution control).
- Type of gas turbine used for producing electricity.

Two approaches were considered for modelling these gasifiers:

1. Using a detailed particulate gasification model, or
2. modelling the formation of products based on equilibrium.

The study focused specifically on coals found in the United Kingdom, with the 5 bituminous coals as listed in table 4.6 selected.

Table 4.6: Coals studied by Norman et al. (1997)

Coal	Ash yield (wt %)
Daw Mill	3.9
Kellingley	4.3
Longannet	19.0
Nadins	9.4
Thoresby	4.2

After numerous experiments, Norman et al. proposed the reactions of table 4.7 to take place during gasification.

Table 4.7: Reactions proposed to occur in gasifier

$2H_2 + O_2$	$\rightarrow$	$2H_2O$
$CO + \frac{1}{2}O_2$	$\rightarrow$	CO_2
$CO + H_2O$	$\rightleftharpoons$	$CO_2 + H_2$
$CH_4 + H_2O$	$\rightleftharpoons$	$CO + 3H_2$
$CH_4 + 2O_2$	$\rightleftharpoons$	$CO_2 + 2H_2O$
$CO + H_2$	$\rightleftharpoons$	$H_2O + C$
$CO + 3H_2$	$\rightleftharpoons$	$H_2O + CH_4$
$CO_2 + 4H_2$	$\rightleftharpoons$	$2H_2O + CH_4$

The actual reactions undergone by these species in a gasifier are complex and no attempt has previously been made to incorporate detailed reaction schemes into any models. The study therefore involved investigating the equilibrium concentrations of the trace elements known to occur in the selected coals.

The following general assumptions were made:

1. The system has reached equilibrium. The controlling effect of the kinetics implies that the composition of the gases will not reach equilibrium at lower temperatures.

2. Condensation of certain elements is not considered. This may lead to compositions being predicted as more than their actual values.

Norman et al. found (using EQUITHERM) that whether using equilibrium modelling or a more detailed particulate model, equilibrium is reached for the main gaseous components.

4.4.2 Modelling of Kinetics

The findings summarised above had to be combined and the significance thereof ascertained, i.e. the modelling of the internal gasifier kinetics had to be explained and summarised. Since most of the research on this topic focused on gasifier effluent composition and temperature, it was decided to keep with the status quo and investigate the reaction kinetics by evaluating the effluent gas composition given a certain temperature and pressure. ASPEN was selected as initial simulation environment and a simplified modelling exercise undertaken.

The model objective could hence be formulated as: "The accurate prediction of the effluent gas composition at specified conditions (temperature and pressure), for a generic gasifier." The system was selected as the entire gasifier, with the system boundaries the physical gasifier walls. The key variables are the mole fractions or flows out of the gasifier.

Since many researchers proved that a kinetics-free model works sufficiently well for specific types of gasifiers, this postulate was investigated for a generic gasifier and with a variety of different coals. For a generic gasifier, the direction and pattern of flow as well as the specific contact between the different phases and stages are neglected and only the reactions thought to take place considered. The reactions proposed by the researchers mentioned before (Kosky & Floess (1980), Norman et al. (1997), Arri & Amundson (1978), Yoon et al. (1976)) were combined and investigated for a range of different coals and operating conditions. There were a few reactions in common, which resulted in the 13 unique reactions found in table 4.8.

Table 4.8: Reactions proposed to occur in gasifier

$$\begin{array}{rcl}
C + H_2O & \rightleftharpoons & CO + H_2 \\
C + CO_2 & \rightleftharpoons & 2CO \\
C + 2H_2 & \rightleftharpoons & CH_4 \\
CO + H_2O & \rightleftharpoons & CO_2 + H_2 \\
C + \frac{1}{2}O_2 & \rightleftharpoons & CO \\
C + O_2 & \rightleftharpoons & CO_2 \\
2H_2 + O_2 & \rightleftharpoons & 2H_2O \\
CO_2 + 4H_2 & \rightleftharpoons & 2H_2O + CH_4 \\
CO + \frac{1}{2}O_2 & \rightleftharpoons & CO_2 \\
CH_4 + H_2O & \rightleftharpoons & CO + 3H_2 \\
CH_4 + 2O_2 & \rightleftharpoons & CO_2 + 2H_2O \\
CO + 3H_2 & \rightleftharpoons & H_2O + CH_4 \\
CO + H_2 & \rightleftharpoons & H_2O + C
\end{array}$$

Kosky & Floess (1980), Norman et al. (1997), Arri & Amundson (1978), Yoon et al. (1976) showed that equilibrium is thought to be reached for many reactions occurring inside gasifiers. Due to the high operating temperatures typically used for gasification, all the proposed reactions were considered to be at equilibrium.

Yoon et al. (1976) also assumed that the heat of reaction generated in the reaction of coal to form char and pyrolysis products is negligible.

Daw Mill coal

Of the coals studied by Norman et al., as listed in table 4.6, the Daw Mill coal was selected for initial investigation. The composition of the Daw Mill coal as reported by Norman et al. is found in table 4.9.

Norman et al. obtained experimental effluent compositions for this coal, using the ABGC gasifier at 770℃, and 10 atm. These experimental values, together with the predicted compositions of the Norman et al. model are displayed in table 4.10. The amount of coal, air and steam fed was calculated from the predicted effluent composition and coal analysis, and can be viewed in Appendix A. This was then simulated in ASPEN using an equilibrium reactor with the conditions and feed rates as reported by Norman et al.. The 13 reactions as proposed were specified and the resulting effluent composition shown in table 4.10. Figure 4.4 is a schematic of the

Table 4.9: Composition of the Daw Mill coal

Component	Weight %	moles
Ash	3.9	n/a
Carbon	76.8	6.4
Hydrogen	4.7	4.7
Nitrogen	1.4	0.1
Sulfur	1.3	0.04
Chlorine	0.2	0.0057

ASPEN worksheet used for this simulation.

Table 4.10: Effluent gasifier composition (mol %)

Component	Experimental	Norman	13 Eq	4 Eq	3 Eq
CH_4	1.5	1.32	1.26	1.26	0
CO	18.5	15.8	18.73	18.73	20.45
CO_2	9.0	6.6	8.81	8.81	7.65
H_2	13.9	14.9	14.37	14.37	16.75
H_2O	5.7	5.5	5.57	5.57	5.16
N_2	51.4	55.7	51.08	51.08	49.82

The 13 equations were systematically reduced to obtain a more concise reaction scheme which still yields the same, or very similar effluent compositions.

It was found that 4 equations (as displayed in table 4.11), predict similar yields to the 13 postulated equations, with the predicted compositions only differing at the 5th or 6th significant number. It is evident that the alternatives using either 13 or 4 equations yield the best results. No combination of three equations gave satisfactory results. The simulation using 4 equations is the simplest alternative which still gives acceptable results. The three equation reaction scheme which yielded the best results (shown in table 4.10), discarded the methane forming reaction, $C + 2H_2 \rightleftharpoons CH_4$.

Norman et al. performed the gasification experiments for a variety of temperatures. It was decided to test the newly proposed 4 equation reaction scheme across this temperature range. The simulation conditions were adjusted appropriately and the results compared to that reported by Norman et al.. See table 4.12.

The proposed reaction scheme is seen to give very accurate predictions for all the components at every temperature point. Two other coals with differing analyses were also simulated and the results discussed hereafter:

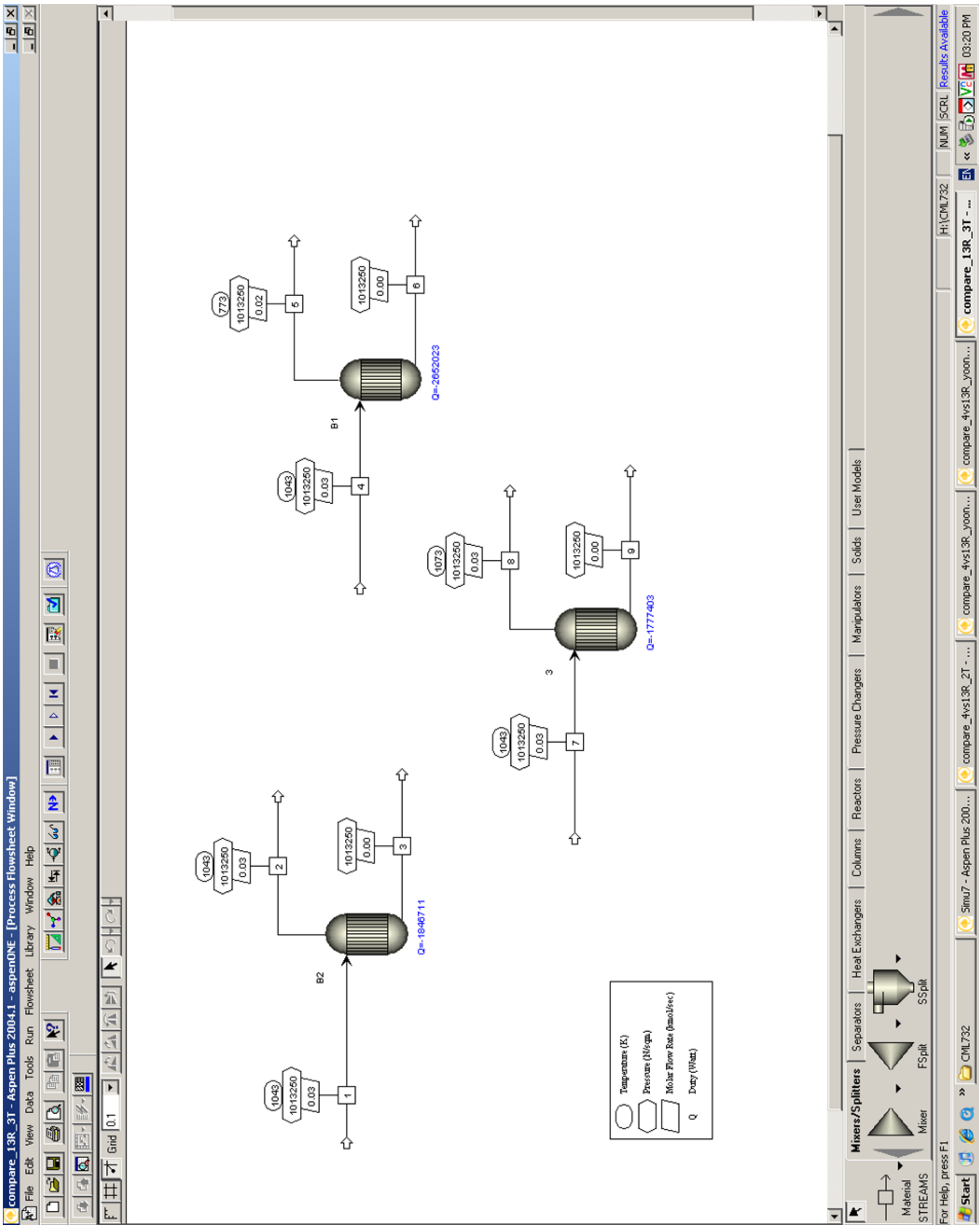


Figure 4.4: Comparing different reaction schemes in the ASPEN environment

Table 4.11: New reaction scheme

$$\begin{aligned} C + H_2O &\rightleftharpoons CO + H_2 \\ C + CO_2 &\rightleftharpoons 2CO \\ C + 2H_2 &\rightleftharpoons CH_4 \\ C + O_2 &\rightleftharpoons CO_2 \end{aligned}$$

Table 4.12: Effluent compositions across temperature range

	Temperature									
Component	500°C		750°C		760°C		770°C		800°C	
	Exp	Sim	Exp	Sim	Exp	Sim	Exp	Sim	Exp	Sim
CH_4	10.1	9.89	2.1	1.74	1.8	1.49	1.5	1.26	0.9	0.73
CO	2.4	2.55	17.6	17.87	18.0	18.32	18.5	18.73	19.6	19.73
CO_2	21.3	21.22	9.7	9.46	9.3	9.12	9.0	8.81	8.1	8.05
H_2	2.3	2.46	13.1	13.68	13.5	14.04	13.9	14.37	14.8	15.10
H_2O	4.0	4.00	5.7	5.52	5.7	5.54	5.7	5.57	5.8	5.67
N_2	59.9	59.67	51.9	51.54	51.7	51.30	51.4	51.08	50.8	50.55

Taiheiyo bituminous

Chen et al. (2000) developed a "comprehensive three-dimensional simulation model" for entrained flow coal gasifiers. They conducted a number of simulations on a 200 ton per day two-stage air blown entrained flow gasifier. This gasifier was specifically developed for the IGCC (Integrated Gasification Combined Cycle) process. Their model subsequently predicted the temperature profile and effluent gas composition. The coal used during their experimental work was the Taiheyo bituminous coal with ultimate analysis as shown in table 4.13.

Table 4.13: Ultimate analysis of the Taiheyo bituminous coal

Component	Weight %	Moles
Carbon	77.6	6.47
Hydrogen	6.5	6.5
Nitrogen	1.13	0.081
Oxygen	13.9	0.869
Sulfur	0.22	0.007

Using this analysis, together with the experimental conditions as reported (2.7 MPa, 900℃), the newly proposed kinetics-free reaction scheme using simply the 4 equations of table 4.11 was used to determine the exit gas composition. This was once again simulated in ASPEN using an equilibrium reactor and the values obtained compared with the experimental values and those of the Chen et al. model (table 4.14).

Table 4.14: Effluent gasifier composition (mol %)

Component	Experimental	Chen	Simulated
CO	23.3	20.3	22.36
H_2	10.1	11.0	7.88
CH_4	0.4	0.0	0.21
CO_2	4.7	6.4	5.76
H_2O	-	2.4	2.57
N_2	61.5	59.9	61.18

Both models are seen to predict the effluent compositions accurately. The 4 equation, equilibrium reaction scheme does however seem to be slightly superior. Refer to Appendix B for detailed calculations.

Illinois low activity coal

Yoon et al. (1979b) performed analyses on Lurgi gasifiers using the U.S. coals from Illinois and Wyoming. The Illinois coal is reported to have low activity, while the Wyoming alternative is thought to be highly active. Their work considered gasification using either air or oxygen. They also derived a model to predict the effluent temperature and gas composition. No mention is made of experimental data in their article. It was therefore decided to test the 4 equation, equilibrium reaction scheme on the same conditions and coal composition as reported (table 4.15) to ascertain whether comparable results can be obtained. Only the results for the Illinois coal using air as feed are reported here (table 4.16).

Table 4.15: Composition of Illinois coal

Component	Molar Ratio	moles
C	1	
O_2	0.357	6.5
H_2O	2.39	0.1
N_2	1.34	0.0057

The experimental conditions are reported as 25 atmospheres and 615°C. ASPEN was again used to perform the simulations.

Table 4.16: Effluent gasifier composition (mol %)

Component	Yoon	Simulation (615°C)	Error	Simulation (720°C)	Error
CO_2	18.4	23.49	5.09	19.95	1.55
CO	12.0	5.16	-6.84	12.12	0.12
H_2	26.6	6.75	-19.85	12.33	-14.27
CH_4	3.7	9.19	5.49	5.24	1.54
Other	0.7	-	-	-	-
N_2	38.6	42.82	4.22	39.97	1.37

Table 4.16 shows that the results of the proposed reaction scheme do not compare well with that of Yoon et al. (1979b). A temperature sensitivity analysis was performed, which showed better comparison when the simulation is performed at 720°C. The value of 615°C, reported by Yoon et al. (1976) is that of the effluent gas, but it is not stated exactly where this measurement was taken. It may be that this is some distance downstream from the gasifier and that this temperature therefore differs significantly from the reaction temperature inside the gasifier. The temperature of 720°C may then in fact be closer to the gasification temperature.

Migration to UNISIM

The postulated kinetics-free, 4-equation, equilibrium, reaction scheme was subsequently also tested in the UNISIM design engine. Devon Clack, a final year Chemical Engineering student performed this work as part of his final year project. He modelled the Lurgi gasifiers employed by Sasol Limited, and compared the effluent compositions calculated by the postulated model with actual data obtained from Sasol (see figure 4.5 for a schematic of the UNISIM process flow diagram used). Clack (2008) found that: "On average, the difference between the simulation and Sasol plant data of the concentrations of CO, CO_2 and CH_4 in the syngas differ by 2.2 mol %, 4.1 mol % and 0.65 mol % correspondingly." He explains the perceived discrepencies:

- "The coal analysis that was used in the simulation was five years older than the dynamic gasifier data, and consequently may not correspond."
- "The operating temperatures specified in the Sasol data were very low, too low for there to be as little methane formation as was reported. It was concluded

that the temperatures must have been taken downstream of the gasifier and were not the operating temperature of the unit. A temperature of 650℃ was used in the simulation."

- "The Sasol data was not well documented; some information had to be assumed. This may have a profound influence on the resulting simulation concentrations."

Clack concluded by stating that: "These results were produced with limited optimization time. The results can definitely be improved upon. Also, considering the feed data obtained from Sasol, one cannot expect excellent results from average data. But nonetheless, a dynamic Lurgi coal gasifier simulation was produced."

Conclusion

It is therefore evident that not only does the kinetics-free, 4-equation, equilibrium reaction scheme accurately predict the effluent gas composition of three different types of gasifiers using three different feed coals (ASPEN simulations), but also successfully calculated effluent compositions for a Sasol gasifier in UNISIM DESIGN. This investigation therefore finds that the reaction kinetics of any gasifier can be well approximated by the 4-equation, kinetics-free, equilibrium reaction scheme as postulated.

4.5 Heat Transfer Effects

In the chemical reactions just considered, the rate of disappearance of a certain species is dependent on temperature and composition according to the "rate law" as defined by Fogler (2002):

$$-r_A = [k_A(T)][f(C_A, C_B, ...)] \tag{4.11}$$

with the temperature dependence of the specific reaction rate, k_A given by the Arrhenius equation:

$$k_A = Ae^{-E/RT} \tag{4.12}$$

Chemical reaction rates are therefore clearly dependent on temperature, with the product equilibrium and extent of reaction influenced by the temperature achieved.

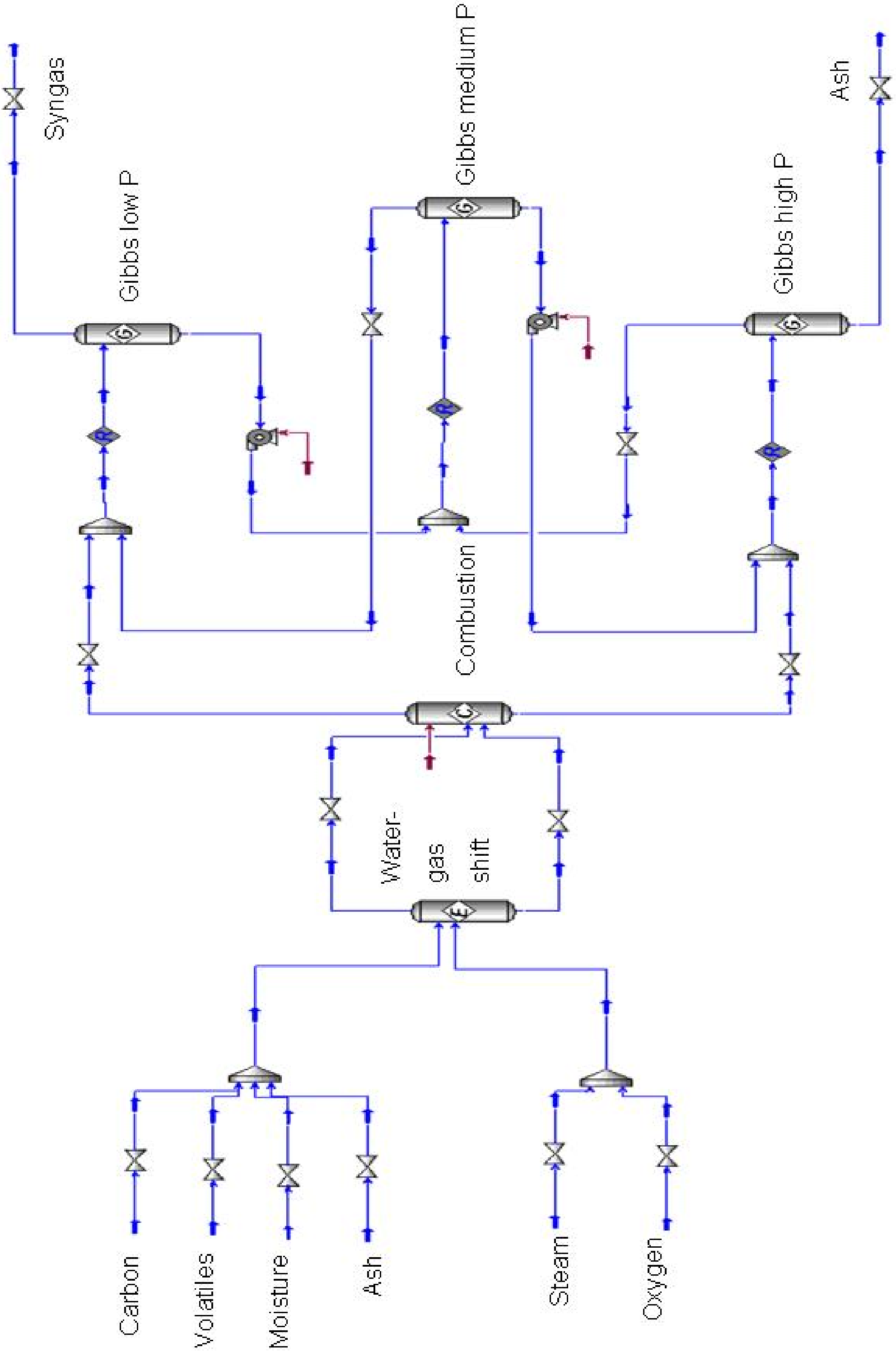
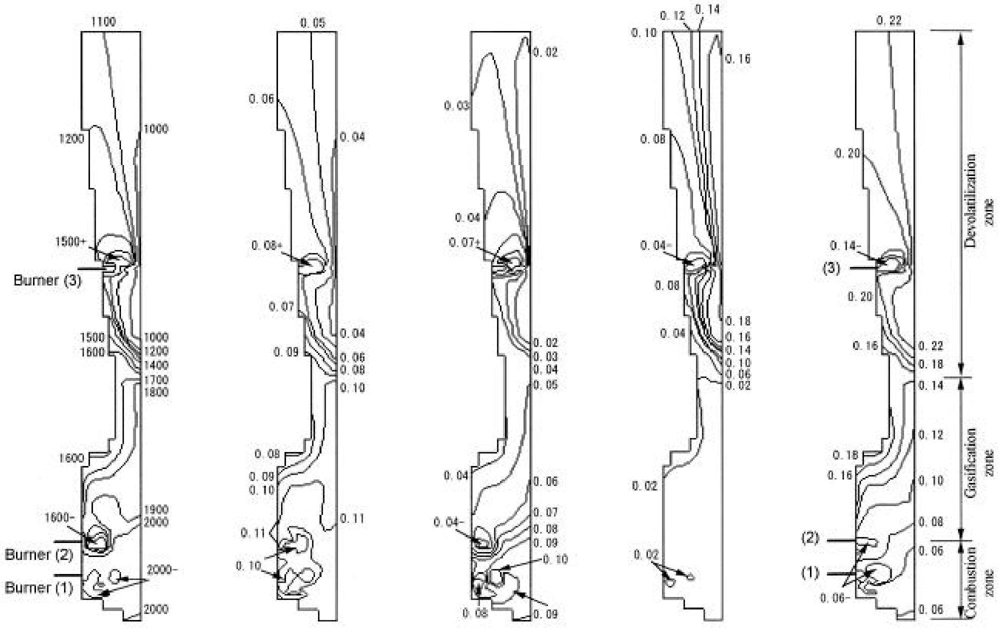


Figure 4.5: Process flow diagram for simulating a Lurgi gasifier in UNISIM (Clack, 2008)

However, gasifiers do not exhibit a single temperature throughout the reacting regions, due to the possibility of several different input streams, entering at various locations and with different temperatures. These streams mix, and a variety of reactions occur before the products leave the gasifiers. Therefore, a temperature profile would be present, rather than a uniform temperature throughout the gasifier length. This deduction has been confirmed by various researchers.

Chen et al. (2000) performed experiments, simulating the distributions of the gas temperature and compositions throughout the gasifier. They found that the gas temperature typically decreased along the height of the gasifier, and is related to the gas composition. See figure 4.6 for the simulated axial and radial distributions of the gas temperature and compositions.



(a) T (K) (b) CO_2 (c) H_2O (d) H_2 (e) CO

Figure 4.6: Axial distributions of the gas temperature and the gas compositions in the gasifier (Chen et al., 2000)

Figure 4.7 displays the results of research conducted by Vicente et al. (2003) and shows the simulated distributions for gas temperatures and compositions in a gasifier. Vicente et al. found that these were typically axially symmetrical, with the gas temperature generally decreasing along the gasifier length as well as being linked with the gas composition, i.e. the high concentrations of CO_2 and H_2O are due to the high temperatures, whereas the high concentrations of CO and H_2 are associated with the lower temperatures.

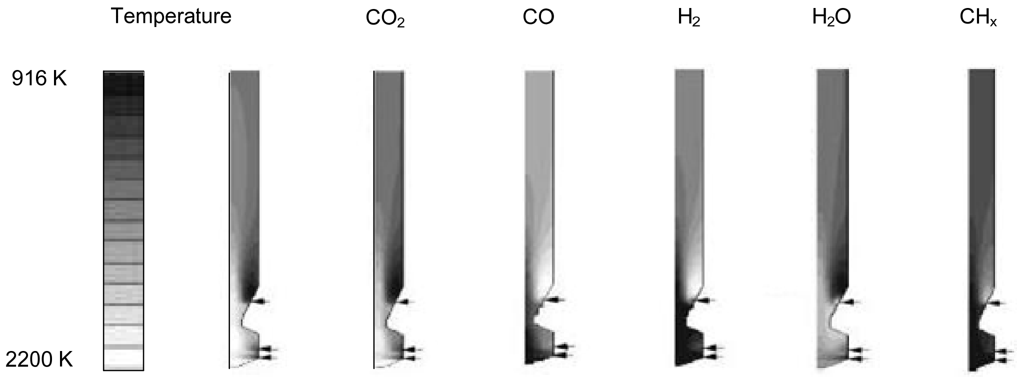


Figure 4.7: Contours of the gas temperature (K) and concentrations of CO_2, CO, H_2, H_2O and CH_x (mass fractions) (Vicente et al., 2003)

Welty et al. (2001) reports that there are three modes of energy transfer: conduction, convection and radiation. All of these are present in some way or another in every heat transfer process.

4.5.1 Conduction

Welty et al. (2001) states that there are two primary ways by which energy is transferred via conduction. The first being that of molecular interaction, where molecules of a higher energy level (i.e. temperature) convey energy to neighboring molecules of a lower energy level. The second method is primarily associated with pure metallic solids and involves "free" electrons. The equation used to describe heat transfer through conduction was proposed by Fourier:

$$\frac{q_x}{Ar} = -k\frac{\mathrm{d}T}{\mathrm{d}x} \tag{4.13}$$

The negative sign of equation 4.13 signifies that the heat flow is in the direction of the negative temperature gradient.

4.5.2 Convection

The second method of heat transfer entails exchange of energy between a surface and an adjoining fluid. The equation describing heat transfer via convection was derived by Newton and is as follows:

$$\frac{q}{Ar} = h\Delta T \tag{4.14}$$

4.5.3 Radiation

The difference between radiant heat transfer and transfer by means of either convection or conduction is that no physical medium is required for the propagation of radiant heat transfer. Heat transfer via radiation is in fact maximum when the energy exchanging surfaces are separated by an ideal vacuum. The rate of energy radiation is expressed by the Stefan-Boltzmann law of thermal radiation:

$$\frac{q_r}{A_r} = \sigma T^4 \tag{4.15}$$

4.5.4 Heat transfer in gasifiers

- Chen et al. (2000) found that convective, conductive and radiative heat transfer was exhibited between the gases, particles as well as reactor walls of gasifiers. They also reported that the scattering properties of these particles to radiation can be assumed to be isotropic.

- The first process undergone by the coal during gasification, drying and volatilization, generally takes places in the upper parts of gasifiers. Yoon et al. (1978) proposed that both the drying and volatilization steps take place immediately and completely, while Kosky & Floess (1980) proposed that the heat of reaction associated with the volatilization step is zero.

  Yoon et al. (1978) suggested that the reaction environment can be divided into two distinct regions – an adiabatic core and a boundary layer. The boundary layer is estimated to be no more than about 10 mm thick (Yoon et al., 1978). The influence of the boundary layer as well as heat loss to the mantle (convection) is therefore generally assumed to be insignificant.

- From figure 4.8, Yoon et al. showed that the radial thermal conductivity of the coal is very low in the reaction chamber.

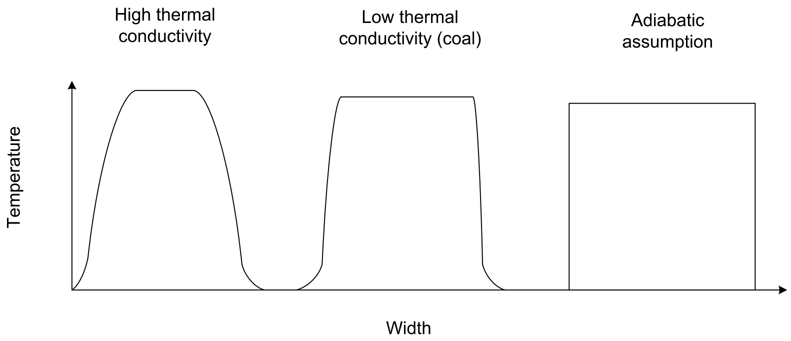


Figure 4.8: Radial thermal conductivity (adapted from Du Plessis (2003))

- Because the solid-gas phase reactions take place inside the solid phase, it is believed that the heat transfer between these phases is high. This leads to the general assumption that the temperatures of the gas and solid are essentially the same Yoon et al. (1978). No temperature gradient is therefore present.

4.6 Mass Transfer Effects

Apart from kinetics and heat transfer, the other major role player in chemical processes is mass transfer. Welty et al. (2001) states that there is a natural tendency for systems that contain components of different concentrations to tend to minimize these concentration differences. This occurs via the transfer of mass from the component of high concentration to the component of lower concentration and is called mass transfer. Mass is transferred by either a random molecular motion occurring in quiescent fluids (molecular mass transfer) or from a surface into a passing fluid, assisted by the dynamic flow characteristics (convective mass transfer).

4.6.1 Molecular Mass Transfer

The laws of mass transfer convey the relationship between the flux of the diffusing component and the concentration gradient causing the transfer of mass. Because mass transfer occurs exclusively in mixtures, the assessment entails an examination of the effects of each component (Welty et al., 2001).

In multicomponent systems, the different species involved will generally travel at different velocities, necessitating the averaging of the velocities of each component in order to evaluate the overall gas mixture velocity. This gives rise to the mass-average velocity for multicomponent mixtures as stated in equation 4.16:

$$v = \frac{\sum \rho_i v_i}{\sum \rho_i} = \frac{\sum \rho_i v_i}{\rho} \tag{4.16}$$

The diffusion velocity of a component is the velocity of that component relative to the mass-average velocity. Fick's law states that a component can only have a relative velocity if gradients in the concentration are present (Welty et al., 2001). Fick's law defines the diffusion of a component in an isobaric, isothermal system according to equation 4.17:

$$J_A = -D_{AB} \nabla_{C_A} \tag{4.17}$$

De Groot offered a more universal relationship, not limited to isobaric and isothermal systems:

$$J_A = -cD_{AB} \nabla_{y_A} \tag{4.18}$$

4.6.2 Convective Mass Transfer

Welty et al. states that convective mass transfer is dependent on the dynamic characteristics, as well as the transport properties of the flowing fluid, and similar to molecular mass transfer, occurs in the direction of decreasing concentration. The generalized convective mass transfer rate equation is:

$$N_A = k_c \Delta c_A \tag{4.19}$$

with the convective mass transfer coefficient, k_c a function of the fluid and flow properties, the system geometry and the concentration difference, Δc_A.

4.6.3 Mass transfer in gasifiers

When studying the various gasifier technologies, it becomes evident that the gas phase residence times are in the order of seconds, while the solid phases generally spend several hours inside the reaction chambers. Due to the faster dynamics of the gas phase, the hold-up of this phase is assumed to be zero as it approaches steady state. This has as effect that the volumetric flow rates of both the inlet and outlet gases can be taken as equal and constant.

When examining the volumetric flow of the solids on the other hand, use is made of the shrinking core model. The shrinking core model describes the consumption of solid particles by either dissolution or reaction, i.e. the material being consumed is "shrinking." This shrinking core model finds application for heterogeneous reactions where a component in the gas phase reacts with a species enclosed in a solid matrix (Fogler, 2002), as shown in figure 4.9. As reaction takes place and material is eliminated from the solid particle, the reactant gas has to diffuse further into the solid to contact the unreacted solid phase. The reaction taking place at the solid surface (equation 4.20), is very fast, causing the rate of oxygen diffusion to the surface to be the rate controlling step.

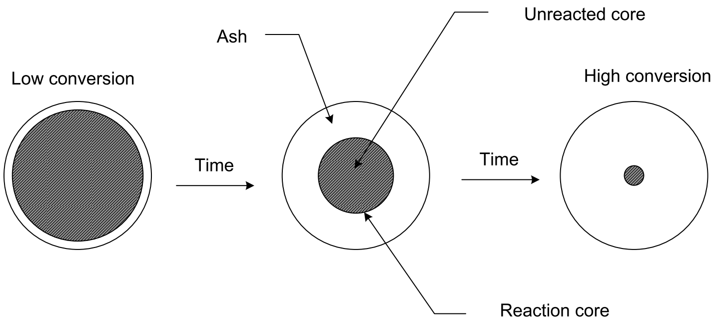
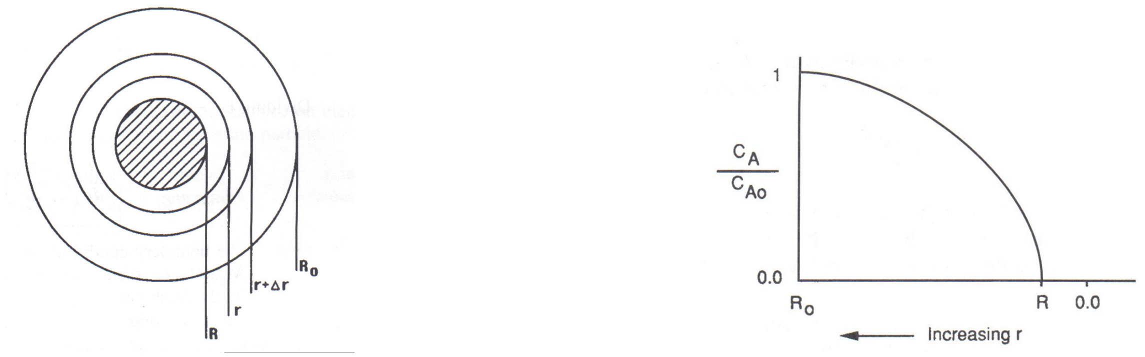


Figure 4.9: Shrinking core model (adapted from Du Plessis (2003))



(a) Solid sphere as modelled by the shrinking core model

(b) Oxygen concentration profile in the pellet

Figure 4.10: Shrinking core model Fogler (2002)

$$C + O_2 \rightarrow CO_2 \tag{4.20}$$

It should be noted that the shrinking core model was derived for combustion processes, but is just as applicable for gasification, with equation 4.20 not necessarily going to completion, but forming CO instead.

Even though the solid core shrinks as time passes (essentially an unsteady state process), Fogler states that it is generally assumed that the concentration profiles at any time step is the steady state profiles over the distance $(R_0 - R)$, as illustrated in figure 4.10. This assumption is called the quasi-steady state assumption (QSSA).

To investigate how the radius of the unreacted carbon changes with time, a number of things should be considered:

- The rate of oxygen diffusion to the carbon surface.
- The rate of carbon consumption.
- For every mole of O_2 that diffuses into the pellet, 1 mole of CO_2 (or 2 moles of CO) diffuses out.

Figure 4.10 shows the oxygen concentration profile as the inner core recedes to a radius R, with r = 0 denoting the center of the spherical pellet.

This model proposes that, even though the density of particles change as a result of reaction, their volume remains constant throughout. This implies that the volume of the reaction chamber stays constant as the various reactions occur.

Chapter 5

Conclusion

As shown in this document there exists many different gasifiers in various stages of development. 16 of these gasifiers situated all over the world were discussed in detail and their characteristics summarised. From this summary the differences and mutual properties among the gasifiers were discussed with special emphasis on their effect on gasifier modelling. A detailed, step-by-step modelling procedure was also provided. Apart from the specific gasifier characteristics, the phenomena of reaction kinetics, heat transfer and mass transfer were also investigated to ascertain their significance on gasifiers and specifically gasifier modelling. It was found that:

- A kinetics-free, 4-equation, equilibrium reaction scheme can be used to accurately predict the effluent gas composition of a generic type gasifier. This was verified using three different types of feed coal and successfully simulated in both ASPEN and UNISIM DESIGN.
- Convective, conductive and radiative heat transfer is exhibited between the gases, particles and reactor walls of gasifiers. The radiation scattering properties of the particles can be assumed to be isotropic.
- Yoon et al. proposed that both the drying and volatilization steps take place immediately and completely, while Kosky & Floess (1980) proposed that the heats of reaction associated with the volatilization step is zero.
- The boundary layer associated with the reacting species is estimated to be no more than about 10 mm thick. The influence of this boundary layer as well as heat loss to the mantle (convection) can therefore generally be assumed to be insignificant.

- The radial thermal conductivity of the coal in the reaction chamber is very low.
- Because the solid-gas phase reactions take place in the solid phase, it is believed that the heat transfer between these phases is high, which leads to the assumption that the temperatures of the gas and solid are essentially the same and that no temperature gradient is present.
- The gas phase residence times are in the order of seconds, while the solid phases generally spend several hours inside the reaction chambers. Due to the faster dynamics of the gas phase, the hold-up of this phase is assumed to be zero as it approaches steady state, which causes that the volumetric flow rates of both the inlet and outlet gases can be taken as equal and constant.
- When examining the volumetric flow of the solids on the other hand, use is made of the shrinking core model. This model proposes that, even though the density of particles change as a result of reaction, their volume remains constant throughout. This implies that the volume of the reaction chamber stays constant as the various reactions occur.

By taking all of this into account, it should be possible to develop a generic gasifier model in any modelling environment.

Chapter 6

Appendices

6.1 Appendix A

The weight percentages as reported for the Daw Mill coal were converted to moles and displayed in table 4.9. Performing a carbon balance on the experimentally obtained effluent analysis (using as base 100 moles of effluent), yields:

Total Carbon = Carbon in CH_4 + Carbon in CO_2 + Carbon in CO

= 1.5 + 18.5 + 9

= 29 moles

Doing this for each element gives:

Element	Moles Out
C	29
H	45.2
O	42.2
N	102.8

This should be the same as the amounts of these elements entering the gasifier (as no matter is created). Coal reacts with steam and oxygen or air. The only source of carbon is the coal. The coal should therefore contain 29 moles of carbon. With the coal analysis as given in table 4.9, the amount of coal fed was calculated as:

29 moles of carbon fed / 6.4 moles of carbon in coal = 4.53 moles of coal fed.

In multiplying the coal analysis as given with this factor, the amount of each element fed to the gasifier (as coal) is obtained:

There is more nitrogen exiting as effluent than entered with the coal (102.8 - 0.453 = 102.35 moles). This suggests that air was used as oxidising agent and the nitrogen

Element	Moles in Coal
C	29
H	21.29
N	0.453
S	0.18
Cl	0.026

entered as such. In general, air is considered to consists of 79 percent nitrogen and 21 percent oxygen, which gives the ratio

$\frac{Nitrogen}{Oxygen} = \frac{79}{21} = 3.762$

The amount of elemental oxygen entering as air is hence calculated as:

Elemental nitrogen as air / 3.762 = 102.35 / 3.762 = 27.21 moles.

The balance of the oxygen will enter as steam:

Oxygen as steam = total oxygen out - oxygen in coal - oxygen in air

= 42.2 - 27.21 = 15 moles

which gives 15 moles of water.

6.2 Appendix B

The weight percentages as reported for the Taiheiyo bituminous coal were converted to moles and displayed in table 4.13. Performing a carbon balance on the experimentally obtained effluent analysis (using as base 100 moles of effluent), yields:

Total Carbon = Carbon in CH_4 + Carbon in CO_2 + Carbon in CO

= 0.4 + 4.7 + 23.3

= 28.4 moles

Doing this for each element gives:

Element	Moles Out
C	28.4
H	21.8
O	32.7
N	123

This should be the same as the amounts of these elements entering the gasifier (as no matter is created). The only source of carbon is the coal, the coal should therefore contain 28.4 moles of carbon. With the coal analysis as given in table 4.13, the amount of coal fed was calculated as:

28.4 moles of carbon fed / 6.47 moles of carbon in coal = 4.39 moles of coal fed.

In multiplying the coal analysis as given with this factor, the amount of each element fed to the gasifier (as coal) is obtained:

Element	Moles in Coal
C	28.4
H	28.5
N	0.354
O	3.815
S	0.03

This does not make sense. It is reported that 21.8 moles of hydrogen exit in the effluent, but the coal itself contains 28.5 moles of hydrogen. There is hence less hydrogen exiting than contained in the coal, this is even without the hydrogen entering as steam! Something had to be done. Seeing that the successful analysis of coal is a very intricate and difficult process, it is not impossible that the coal composition as reported is faulty. It is also highly unlikely that the coal composition would be uniform throughout. The analysis was perhaps done on a hydrogen deprived part of the coal. It was therefore assumed that the effluent composition was analysed accurately and the coal composition adjusted to satisfy the mole balances.

There is more nitrogen exiting as effluent than entered with the coal (123 - 0.354 = 122.65 moles), which suggests that air was used as oxidising agent and the nitrogen entered as such.

The amount of elemental oxygen entering as air is calculated as:

Elemental nitrogen as air / 3.762 = 122.65 / 3.762 = 32.6 moles, which is added to the oxygen reported to be in the coal to give the overall elemental oxygen entering.

The revised elemental quantities entering the gasifier is then as follows:

Element	Moles in Coal
C	28.4
H	28.5
N	122.65
O	36.42
S	0.03

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