

## Thermal Conversion of Biomass, Pyrolysis and Gasification: A Review

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### Abstract

*Paper reviews the state of the art of thermochemical conversion of biomass. Thermochemical conversion technologies include combustion, gasification and pyrolysis. The dominant biomass conversion technology will be gasification, as the gases from biomass gasification are intermediates in the high-efficient power production or the synthesis from chemicals and fuels. In the use of gases from biomass gasification it is important to understand that depending on the gasification temperature a syngas (>1200°C) or a product gas (<1000°C) is generated, of which the latter can be converted into syngas by catalytic or thermal cracking. The Entrained flow gasification is the most suitable technology for high temperature gasification. The characteristics of the generic types of thermochemical conversion – gasification and pyrolysis - are described. The range of products derivable from each system are discussed and related to each technology.*

**Keywords** – Thermal conversion, Gasification, Pyrolysis, Biomass, Syngas.

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### I. INTRODUCTION

Biomass gasification is a promising technology, which can contribute to develop future energy systems which are efficient and environmentally friendly in order to increase the share of renewable energy for heating, electricity, transport fuels and higher applications. The process of gasification to produce combustible gas also known as syngas or producer gas from organic feeds was used in blast furnaces over 180 years ago. The possibility of using this gas for heating and power generation was soon realized and then emerged in Europe producer gas systems, which used charcoal and peat as feed material. At the turn of the century petroleum gained wider use as a fuel, but during both world wars and particularly World War II, shortage in petroleum supplies led to widespread re-introduction of gasification. By 1945 the gas was being used to power trucks, buses and agricultural and industrial machines. It is estimated that there were close to 9000,000 vehicles running on producer gas all over the world [1]. After World War II the lack of strategic impetus and the availability of cheap fossil fuels led to general decline in the producer gas industry. However Sweden continued to work on producer gas technology. A decision was then made to include gasifiers in Swedish strategic emergency plans. The contemporary interest in small scale gasifier R&D, for most part dates from 1973 oil crisis. The gasification of carbon-containing materials to produce combustible gas is an established technology. Biomass gasification is a thermochemical process that produces relatively clean and combustible gas through pyrolytic and reforming reactions. The syngas generated can be an important resource suitable for direct combustion, application in prime movers such as engines and turbines, or for the production of synthetic natural gas (SNG) and transportation fuels e.g. Fischer-Tropsch diesel.

For energy production, the major concerns about syngas are its heating value, composition and possible contamination [2]. The proportion of the combustible gas hydrogen (H<sub>2</sub>), methane (CH<sub>4</sub>), carbon monoxide (CO) and moisture determines the heating value of the gas. The composition of syngas depends on the biomass properties and gasifier operating conditions. For a specific gasification system, operating conditions play a vital role in all aspects of biomass gasification. These include carbon conversion, syngas composition, tars and soot formation and oxidation [3]. The main hurdles for large-scale implementation of energy production from solid biomass are the nature of biomass - non uniform, low-energy density, sometimes large ash content - together with the usual inconsistency between the local availability of biomass and the demand for biomass related products: heat, electricity, fuels and chemicals. Usually, import/transport of fossil fuels is cheaper.

The chemistry of biomass gasification is very complex. Biomass gasification proceeds via a two-step process; pyrolysis followed by gasification that includes gas and tars reforming plus char and soot conversion. Pyrolysis is the decomposition of the biomass by heat. This step, also known as devolatilization, is endothermic and produces mainly volatile materials in the form of gaseous and condensable hydrocarbons called tars. The remaining nonvolatile material, containing mainly the carbon material, is referred to as char. The volatile hydrocarbons and char are subsequently converted to syngas in the presence of steam in the second step called gasification.

## **II. PYROLYSIS**

Pyrolysis is a thermal decomposition process that takes place in the absence of oxygen to convert biomass into solid charcoal, liquid (bio-oil) and gas. Pyrolysis is considered to be an industrial realized process for biomass conversion [4-6]. Each component of lignocellulosic (cellulose, hemicelluloses and lignin) biomass is pyrolysed at different rates by different mechanisms and pathways. Lignin decomposes over a wider temperature range compared to cellulose and hemicelluloses which rapidly degrade over narrower temperature ranges. Hence there is an apparent thermal stability of lignin during pyrolysis. Thermogravimetry analysis (TGA) testing of biomass shows that there are three stages for a typical biomass pyrolysis process [4]. The first stage, pre-pyrolysis, occurs between 120 and 200°C with a slight weight loss, when some internal rearrangements, such as bond breakage, the appearance of free radicals and the formation of carbonyl groups take place, with a corresponding release of small amounts of water (H<sub>2</sub>O), carbon monoxide (CO) and CO<sub>2</sub>. The second-stage is the main pyrolysis process, during which solid decomposition occurs, accompanied by a significant weight loss from the initial biomass. The last stage is the continuous char devolatilization, caused by the further cleavage of C-H and C-O bonds. Depending on the reaction temperature, heating rate and residence time, pyrolysis can be classified into slow pyrolysis (intermediate pyrolysis) fast pyrolysis and flash pyrolysis.

### **2.1 Slow Pyrolysis**

Slow pyrolysis has been applied for thousands of years and has been mainly used for the production of charcoal [7-10]. In slow pyrolysis, biomass was typically heated about 500 °C at slow heating rates (up to 10-20°C/min). The vapour residence time varies from 5 min to 30 min [11]. Thus, the components in the vapour phase continue to react with each other, as solid char and liquid are being formed. The main product, charcoal, can be used in a wide range of areas, from domestic cooking and heating to metallurgical or chemical use as the raw material for production of chemicals, activated carbon, fireworks, absorbents, soil conditioners and pharmaceuticals [12]. As reported by Mok et al. [13], a higher yield of charcoal can be obtained from biomass feedstocks with higher lignin contents and lower hemicelluloses contents. In contrast to fast pyrolysis, slow pyrolysis does not necessarily require fine feedstock particle size (smaller than 1 mm).

### **2.2 Fast pyrolysis**

Fast pyrolysis is a process in which very high heat flux are imposed to biomass particles, leading to very high heating rates, in the absence of oxygen. Biomass decomposes to generate vapours, aerosol and char. After cooling and condensation of the vapours and aerosol, a dark brown mobile liquid is formed which has a heating value of about half of the conventional fuel oil. Fast pyrolysis process produces 60-75 wt% of liquid bio-oil, 15-25-wt% of solid char and 10-20-wt% of non condensable gas, depending on the feedstock used. No waste is generated because the bio-oil and solid char can each be used as a fuel and the gas can be recycled back in the process. Fast pyrolysis uses much higher heating rates than slow pyrolysis. While slow pyrolysis is related to the traditional pyrolysis processes for making charcoal, fast pyrolysis is an advanced process which is carefully controlled to give high yields of liquid. Research has shown that maximum liquid yields are obtained with high heating rates, at reaction temperatures around 500°C and with short vapour residence times to minimize secondary reactions. Very short residence times result in incomplete depolymerization of the lignin due to random bond cleavage and inter-reaction of the lignin macromolecule resulting in a less homogenous liquid product, while longer residence times can cause secondary cracking of the primary products, reducing yield and adversely affecting bio-oil properties [14].

The essential features of fast pyrolysis process are:

- very high heating and heat transfer rates, which usually requires a finely ground biomass feed: <1mm;
- carefully controlled pyrolysis reaction temperature of around 500°C in the vapour phase, with short vapour residence times of typically less than 2 s;
- rapid cooling of the pyrolysis vapours to give the bio-oil product.

### **2.3 Flash pyrolysis**

Very fast pyrolysis is sometimes referred to as 'flash pyrolysis' [15], usually in the context of laboratory studies involving rapid movement of substrate through a heated tube under gravity or in a gas flow. Higher

temperatures and shorter residence times than fast pyrolysis are used; the main product distributions are similar to fast pyrolysis. The distinction between flash and fast pyrolysis has largely disappeared and now the term 'flash' has largely disappeared and is gradually being replaced by a more generalized definition for fast pyrolysis.

## **2.4 Fast pyrolysis reactor configuration**

Fast pyrolysis conversion technology has led to design of original reactor systems that provide the essential ingredients of high heating rates, moderate temperatures and short vapour product residence times for liquids. The most commonly used reactors for fast pyrolysis are bubbling fluidized-bed, circulating fluidized-bed, ablative, entrained flow, rotating cone and vacuum reactors. There are three main methods achieving fast pyrolysis.

- Ablative pyrolysis [16-21], in which heat is transferred by conduction: wood is pressed against a heated surface and rapidly moved during which the wood melts at the heated surface and leaves an oil film behind which evaporates. This process uses large particles of wood and is typically limited by the rate of heat supply to the reactor. It leads to compact and intensive reactors that do not need a carrier gas, but with the penalties of surface area controlled system and moving parts at high temperature.

- Bubbling fluidized bed [11] and circulating fluidized bed pyrolysis [22], in which heat is transferred from a heat source to the biomass by a mixture of convection and radiation. The heat transfer limitation is within the particle, thus, requiring very small particles of typically no more than 3 mm to obtain good liquid yields. Substantial carrier gas is needed for fluidisation or transport.

- Vacuum pyrolysis [11], which has slow heating rates but removes pyrolysis products as rapidly as in the previous methods, which thus simulates fast pyrolysis. Larger particles can be accepted but the vacuum leads to larger equipment and higher costs. Total liquid yields are typically lower than 60-65 wt% compared to 75-80 wt% with the previous two methods.

## **2.5 Pyrolysis products**

### **2.5.1 Char**

Char is a porous carbon structure that remains after the hydrogen and oxygen fractions have left the fuel. Char is often defined as the solid residue after pyrolysis. It is often polluted with other components: mineral fractions and after incomplete pyrolysis, large fractions of hydrogen and oxygen, that can still be present in char. Char is believed to contribute to the formation of polycyclic aromatic hydrocarbon (PAHs) during biomass pyrolysis, particularly at low temperature. Char can be used as a fuel in form of briquettes or as a char-oil, char-water slurry; alternatively char can be upgraded to activated carbon and used in purification processes [23]. The properties of the char obtained after biomass pyrolysis have a direct influence on the subsequent char oxidation step, since the amount and type of pores determine the gas accessibility to the active surface sites. Properties of char are decisively affected not only by properties of parent material but also by pyrolysis operating conditions, mainly the heating rate, the maximum temperature and the residence time at this temperature.

### **2.5.2 Pyrolysis liquid**

The liquid product from biomass pyrolysis is known as bio-oil. Bio-oil is not a product of thermodynamic equilibrium during pyrolysis but, it is produced with short residence times and rapid cooling or quenching from the pyrolysis temperature. This condensate is not at thermodynamic equilibrium at storage temperatures. Hence the bio-oil chemical composition tends to change toward thermodynamic equilibrium during storage.

### **2.5.3 Gas**

The third main product from pyrolysis is gas. The gas mainly consists of H<sub>2</sub>, CO<sub>2</sub>, CO and CH<sub>4</sub> together with traces of C<sub>2</sub> species. CO<sub>2</sub> and CO are preferentially produced at low temperature, while H<sub>2</sub> is produced at high temperature.

## **2.6 Pyrolysis reaction schemes**

The exact pyrolysis mechanisms of biomass are still unknown, although substantial literature sources are available on biomass devolatilization kinetics and mechanisms. Many biomass devolatilization models have been developed. One-step global mechanisms and semi-global multi-step mechanisms can be basically distinguished. The simplified approaches define devolatilization rates with single or two-step Arrhenius reaction schemes involving pseudo-species.

The one-step global mechanisms can be shown as:



The reaction kinetic rate ( $k$ ) is expressed in single-step Arrhenius law form as  $k = A \exp(-E_a/RT)$  and the devolatilization rate is expressed as:

$$\frac{-dm_p}{dt} = k[m_p - (1 - f_{v,0})m_{p,0}] \quad (2)$$

where  $m_p$  is the biomass particle mass,  $m_{p,0}$  is the initial particle mass and  $f_{v,0}$  is the initial volatile fraction. For two-step Arrhenius reaction schemes, the kinetic devolatilization rate expressions of the form proposed by Kobayashi [24] are:

$$k_1 = A_1 \exp(-E_a/RT) \quad (3)$$

$$k_2 = A_2 \exp(-E_a/RT) \quad (4)$$

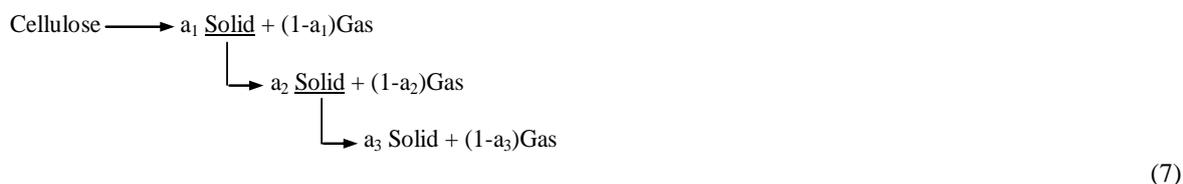
where  $k_1$  and  $k_2$  are competing rates that may control the devolatilization over different temperature ranges. One-step multi-reaction schemes have been developed by Thunman and Leckner [25] via three parallel reactions into char, tar and gas and can be shown as follow:



More recent models are adapted to be able to handle different feedstocks. The composition of the feedstock is represented using three model species. In these models the source species like wood or biomass is replaced by a mixture of cellulose, hemicelluloses and lignin. Most models are only proposed for cellulose, but several schemes were also proposed for the other biomass main components. Several other kinetic schemes for cellulose pyrolysis are proposed below. Broido and Shafizadeh [26] developed a multistep model. In this scheme, cellulose is firstly converted to an active state by an initiation reaction. Hereafter the active cellulose reacts via two competing reactions to produce tar, char and gas.



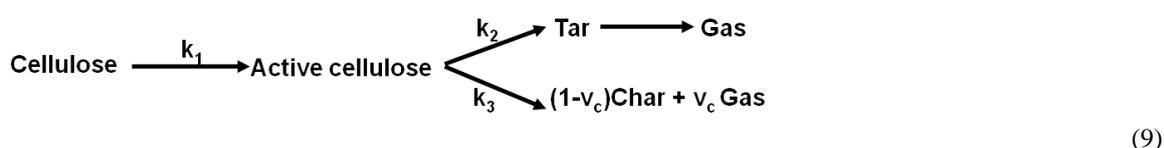
The two competing reactions represent two pathways that the decomposition of cellulose can follow. The “char path” is favored at low temperature (200-280°C) whereas the “tar path” dominates at elevated temperatures (280-340°C). Over the years the validity of the Broido-Shafizadeh model has been disputed by several authors. The use of the initiation step was disputed by Vargyi and Jakab [27] and by Antal and Varhegyi [28] who claimed that in most cases experimental data could be better modeled without this step. A different approach was proposed by Alves [29, 30] and Shrivastava [31]. Their model describes the decomposition by three or two consecutive reactions. This approach requires preset yields for each reaction and therefore lacks of predictive capabilities.



A third approach was the Di Blasi model [32]. It is similar to the model of Thunman. In this model cellulose decomposes via three competing reactions into gas, char and tar. Consecutively the tar is converted by two secondary reactions into secondary gas and char.



The last model discussed here is the Miller-Bellan model [33]. This model uses an adapted form of the Broido-Shafizadeh model. It uses three model species to model biomass decomposition and has three sets of kinetic constants for each model component.



It also uses a secondary reaction for the conversion of tar to smaller gas species. In this reaction no difference is made concerning the origin of the tar.

### 2.7 Secondary reactions

Tars produced during the decomposition of the virgin fuel can decompose further. Several authors suggest a transformation of tar to char and gas following two independent reactions. Tar decomposition is suggested to be catalyzed by the solid matrix of the fuel resulting in char creation on the pore walls.



It is unclear however how this takes place. Among several others, Miller and Bellan [33] suggest a single tar reaction with only gas as product species.



## III. GASIFICATION

### 3.1 Gasification chemistry

Gasification can be seen as an extension of pyrolysis [34-36]; biomass gasification is a complex thermochemical process involving numerous different reactions. The biomass gasification process can be divided into two parts: pyrolysis and gasification. Pyrolysis was discussed in the previous section. In the gasification part the gas, tar and char react further. Gasification, as a core technology for the production of chemicals and clean power, refers to a process converting biomass into either fuel gas (containing CH<sub>4</sub> and some N<sub>2</sub> usually) or syngas (containing mainly H<sub>2</sub> and CO).

During gasification several reactions take place. A simplified representation of these reactions is given in equations (12)-(18) [36-38].

Heterogeneous reactions





$$\Delta H_{298}^0 = 173 \text{ kJmol}^{-1}$$



$$\Delta H_{298}^0 = -75 \text{ kJmol}^{-1}$$

Homogeneous reactions



$$\Delta H_{298}^0 = -41 \text{ kJmol}^{-1}$$

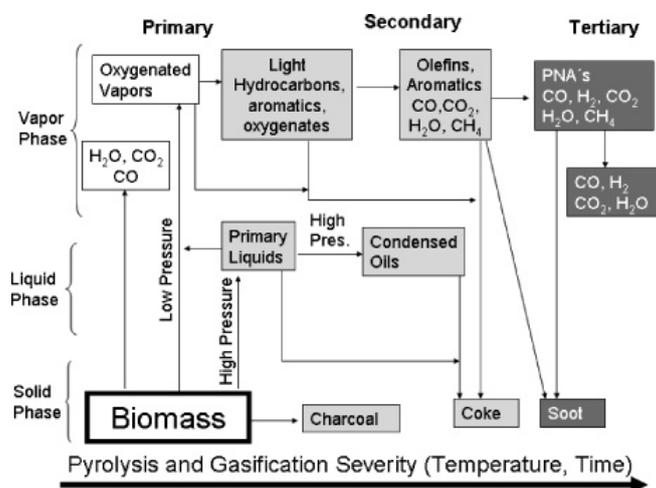


$$\Delta H_{298}^0 = 206 \text{ kJmol}^{-1}$$

Cracking reaction



Evans and Milne [39] observed three major reaction regimes during the gasification process identified as primary, secondary and tertiary regimes as shown in Figure 1. Biomass is mainly converted into a mixture of hydrogen, carbon monoxide, light hydrocarbons, such as methane and other non combustible gas such as carbon dioxide, water vapour and nitrogen. In most cases, the product gas can contain particulates such as char, ash, soot, etc. Reactor temperature is one of the most important operating conditions which affect both the heating value and producer gas composition. The gasification process is usually performed with aid of a gasification agent. The gasification agent can be steam, or air, or enriched air, or oxygen, or a combination of steam and an oxygen source, or carbon dioxide. The process is performed at relative high temperature 600- 1500°C. There are also small amounts of impurities in the gas: char, soot, tars, alkalis, nitrogen compounds, sulphur compounds and chlorine compounds. The composition of the product gas can vary significantly depending on operating conditions (e.g temperature, operating pressure and oxidant agent), type of feed stock, moisture content in the fuel, mode of bringing the reactants into contact inside the gasifier [40]. Moreover, the quality of the product gas is associated with other factors such as the type of gasifier, the residence time and the heating rate which is usually associated with the type of feed stock, particle size and temperature.



**Figure 1.** Gasification and pyrolysis reaction pathways adapted from Milne et al [41]

There are hundreds of different types of gasifiers in the patent literature. However, they can be divided into four main types: Updraft gasifier, Downdraft gasifier, Fluidized-bed gasifier and Entrained flow gasifier.

### 3.2 Gasifier types

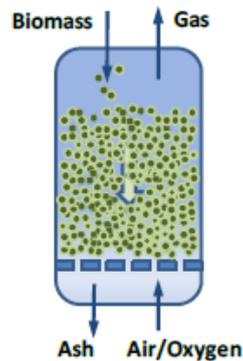
Until now, most gasifiers have been developed and commercialized for the production of heat and power from syngas. The main differences between gasifiers are:

- The type of injection: biomass is either fed from the top of the gasifier, or from the side and then is moved around either by gravity or air flows;
- The gasification agent used;
- The type of heating: it can be done either by partial combustion of the biomass in the gasifier (directly heated), or from an external source (indirectly heated), such as circulation of an inert material;
- The temperature range;

- The pressure range under which the gasifier is operated.

### 3.2.1 Updraft gasifier

Updraft reactor is the simplest and oldest form of gasifier, also known as counter-current gasifier. Biomass enters at the top of the reactor and air/oxygen/steam enters at the bottom of the reactor, flows upward and the product gas leaves at the top. In Figure 2, a schematic representation of an updraft gasifier is given. Complete combustion of char takes place at the bottom of the bed, liberating CO<sub>2</sub> and H<sub>2</sub>O. This hot gas (~1000°C) passes through the bed above, where it is reduced to H<sub>2</sub> and CO and cooled to 750°C. Going up the reactor, the reducing gas (H<sub>2</sub> and CO) pyrolyses the descending dry biomass and finally dries the incoming wet biomass, leaving the reactor at a quite low temperature (~500°C) [42-44].



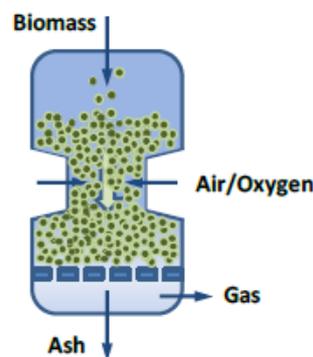
**Figure 2.** The Updraft gasifier (counter-current reactor) scheme [45]

The advantages of updraft gasifiers are that they are a mature technology for heat production, they can be used for small-scale applications, they can handle feedstocks with high moisture content and there is very few carbon in the remaining ash. The disadvantages of updraft gasifiers are that they have a feedstocks size limit, high tar yields which implies extensive syngas cleanup before engine, turbine or synthesis applications.

### 3.2.2 Downdraft gasifier

This gasifier was widespread during World War II. It was used to convert biomass or charcoal into fuel for gasoline vehicles in periods of great shortages of transportation fuels. The downdraft gasifier is a co-current reactor, where the fuel and the gasification agent move in the same direction. In Figure 3, a schematic representation of a downdraft gasifier is given. There are different reaction zones in a downdraft gasifier. In the drying zone, moisture is evaporated from biomass as it slowly moves down towards the pyrolysis zone. In the pyrolysis zone, biomass is converted into char, tars and gas. Some of the pyrolysis products are combusted. Due to the high temperature, tars are cracked. As a result, the produced gas is relatively clean. However, downdraft gasifier has some drawbacks:

- It requires feedstocks drying to low moisture content;
- Syngas exiting the reactor is at high temperature, requiring secondary heat recovery system.



**Figure 3.** The downdraft gasifier (co-current reactor) scheme [45]

### 3.2.3 Fluidized-bed gasifier

The biomass, which is previously reduced to a fine particle size and air, steam, or oxygen enter at the bottom of the reactor. An inert or catalytic bed material is used to transport heat and mass through the reactor. The bed is fluidized by blowing the gasification agent through the bed, which lifts the bed against gravity. As a result, the turbulence in the bed creates an even temperature distribution in the bed. In Figure 4, a schematic representation is given of a fluidized-bed gasifier. Therefore, contrary to fixed bed gasifiers there are no different reaction zones in fluidized bed gasifiers. The operating temperature of a fluidized bed is usually of 700-900°C and the pressure range is between 0 and 70 bar. The most common types of fluidized beds are the bubbling fluidized bed (BFB) and the circulating fluidized bed (CFB).

The advantages of fluidized-bed gasifiers are:

- Exhibit a nearly uniform temperature distribution throughout the reactor;
- Provide high rates of heat transfer between inert material, biomass and gas;
- High conversion possible with low tar and low unconverted carbon.

The disadvantages of fluidized-bed gasifiers are:

- The tolerance of high moisture content feedstocks is much more limited.
- The system design is also more complex, requiring blowers to inject the oxidant at the bottom of the fluidized bed.
- Bed sintering when a biomass with high ash content is used. The alkali components in the ash have the tendency to form low-melting eutectics with silica being the most common bed material, which can lead to loss of fluidization.

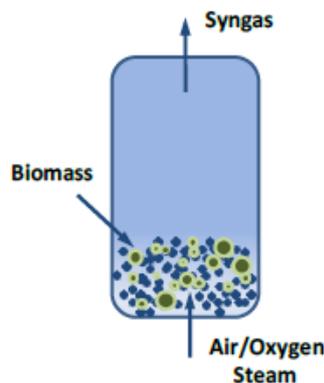


Figure 4. Fluidized-bed gasifiers scheme [45]

### 3.2.4 Entrained flow gasifier

In an entrained flow gasifier, as shown in Figure 5, the biomass is present in dust form. Contact surfaces are very high and hence reaction times are very short. The feed and air move co-currently and the reactions occur in a dense cloud of particles at very high temperature  $>1000^{\circ}\text{C}$  so that firstly the syngas results almost free from tars and secondly ash melts, being then collected at the bottom of the reactor in the form of slag. Conversion in entrained flow reactors is close to 100%. There is little experience with biomass in such systems.

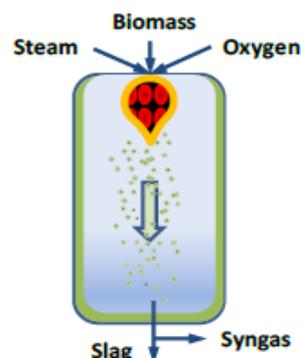


Figure 5. Entrained flow gasifier schematic [45]

### 3.3 Syngas?

Syngas is always confused with the producer gas, however there is a difference between those two terms:

- Producer gas is the mixture of gas produced by the gasification of biomass at relatively low temperatures (700 to 1000°C). Producer gas is composed of carbon monoxide (CO), hydrogen (H<sub>2</sub>), carbon dioxide (CO<sub>2</sub>) and typically a range of hydrocarbons such as methane (CH<sub>4</sub>). Producer gas can be burned as a fuel gas such as in a boiler for heat or in an internal combustion gas engine for electricity generation or combined heat and power. The composition of the gas can be modified by change in of gasification parameters.

- Syngas (synthesis gas) is a mixture of carbon monoxide (CO) and hydrogen (H<sub>2</sub>), which is the product of high temperature steam gasification of biomass. Following clean-up to remove any impurities such as tars, syngas can be used to produce organic molecules such as synthetic natural gas or liquid biofuels such as synthetic diesel or other products as explained below.

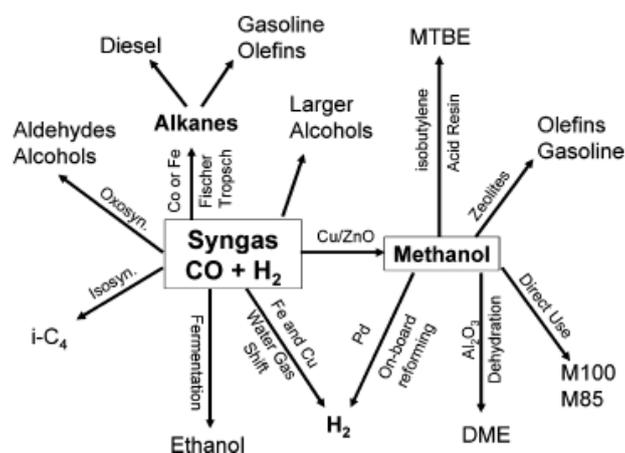
There are four main uses of syngas that are currently being explored for production of liquid fuels:

- Fischer-Tropsch synthesis, a chemical catalytic process that has been used since the 1920s to produce liquid fuels from coal-derived syngas and natural gas;

- Mixed alcohols synthesis, a chemical catalytic process that produces a mixture of methanol, ethanol, propanol, butanol and smaller amounts of heavier alcohols;

- Syngas fermentation, a biological process that uses anaerobic microorganisms to ferment the syngas to produce ethanol or other chemicals;

- Methanol synthesis, also a chemical catalytic process currently used to produce methanol. Figure 6 shows routes for transportation fuels and chemicals production from syngas [46].



**Figure 6.** Pathways for fuel production from syngas adapted from Spath and Dayton [46]

For energy production, the major concerns about syngas are its heating value, composition and possible contamination [2]. The composition of syngas depends on the biomass properties and gasifier operating conditions. The principle contaminants in syngas, as it leaves the gasifier, are: soot, ash, tars, H<sub>2</sub>S (hydrogen sulphide) depending on the sulphur content of the feed and trace quantities of NH<sub>3</sub> (ammonia), COS (carbonyl sulphide), HCl (hydrogen chloride), HCN (hydrogen cyanide), and heavy metals.

## IV. CONCLUSION

Of the variety of technologies available, thermochemical processing has attracted particular attention for converting biomass into more useful and valuable energy products. As pyrolysis and gasification, this technology has been researched and developed for the economic production of fuel products. Gasification is an energy process that produces a gas which can be used to substitute fossil fuels and to reduce CO<sub>2</sub> emission. It is a process which enhances biomass' usefulness as a renewable resource as the product gas (syngas) can easily be transported along existing gas pipelines, and can either be mixed with, or used as a replacement for natural gas combustion in gas turbines for power and heat generation. Syngas can also be used converted to various chemicals such as ethanol, hydrogen, methanol and Fischer-Tropsch fuels.

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