Coal and Biomass Pyrolysis and Gasification – A Review

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Authors' contributions

This work was carried out in collaboration between both authors. Authors NTW and AS both worked on gathering the necessary materials and analyzing them. While author NTW focused on coal-related analysis, author AS instead, worked more on biomass. Both authors have contributed for the contrast analysis and technological progress parts. Both authors read and approved the final manuscript.

Review Article

ABSTRACT

In order to advance the research on using new techniques to produce clean and affordable energy, many gasification experiments have been conducted on different feedstocks and the results have been analyzed. Two promising feedstocks which in the future could help to meet the energy demand are coal and biomass. In this paper, we have collected deep insightful information about the processes of coal and biomass gasification and then compared them. The information contained in this paper cover coal and biomass properties, the devolatization process and the reactions occurring during char gasification. The contrast analysis which has been conducted showed for instance that biomass' higher atomic ratio (H/C, O/C) was the cause of its lower heating value in comparison to coal; carbon conversion values for biomass was significantly higher compared to coal, and during the gasification process, coal feedstock released significantly

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higher pollutant gases (like H_2S) than biomass. In the end, we also presented some innovative technologies that are now developed in the gasification field and which have proven to be highly efficient.

1. INTRODUCTION

With the world's continuous growing economy and population, especially in developing countries, the need for more resources and energy to sustain industrial growth, infrastructures development, food production or climate change issues, has become a challenging matter which should be addressed with appropriate and efficient methods.

One solution would be the production of energy through gasification which is a form of choked combustion or incomplete combustion. It consists of burning solid fuels like wood or coal with a limited amount of air (which otherwise would have led to combustion) and therefore the output gas would still have some combustion potential. The unburned gas is then piped away to burn elsewhere as needed [1]. That can be observed when the material reacts with a certain amount of oxygen (or steam) at high temperatures. The resulting gas mixture is called syngas (from synthesis gas) or producer gas and is a fuel. One real advantage of gasification over combustion is that the use of syngas for that process yields to a better outcome because it involves burning the feedstock at a higher temperature, so that the thermodynamic upper limit to the efficiency as defined by Carnot's rule is higher or (in case of fuel cells) not applicable [2]. Gasification is actually a bit more than just the choked combustion as said earlier. It is more accurately understood as the reaction of solid fuels with air, oxygen, steam, carbon dioxide, or a mixture of these gases at a temperature exceeding 700 °C, which yields to a gaseous product suitable to be used either as a source of energy or as a raw material for the synthesis of chemicals, liquid fuels or other gaseous fuels. It is a series of distinct thermal events put together so as to purposely convert solid organic matters into specific hydrocarbon gases as output.

There is a great variety of feedstocks in use for gasification, each presenting different characteristics, such as the size, shape, bulk density, moisture content, energy content, chemical composition, ash fusion characteristics, and degree of homogeneity. However, the most common feedstocks in many large gasification plants are coal and petroleum coke. Additionally, a variety of biomass and waste-derived feedstocks can be subjected to gasification, namely waste wood, plastics and aluminum, Municipal Solid Waste (MSW), Refuse-derived fuel (RDF), agricultural and industrial wastes, sewage sludge, switchgrass, discarded seed corn, corn stover and other crop residues [3]. The key to improving the adaptability of gasification technology for any feedstock lies in researching and developing feedstock preprocessing technologies and gasifier design.

The focus in this paper would be to conduct a comparative analysis on coal and biomass gasification regarding various aspects of the process. Thus, we will analyze the key differences between coal and biomass gasification during pyrolysis and char combustion process; we will then look at the similitudes and differences existing between the two processes especially the atomic ratio, the char characterization and the pollutant gas emission and finally we will talk about technological progress in the field of gasification.

2. PROPERTIES OF COAL AND BIOMASS

2.1 Properties of Coal

2.1.1 Proximate analysis

Proximate analysis of coal will be used to determine the proportion of fixed carbon, Ash, Volatiles and water contained in a coal sample. These characteristics will directly influence coal properties such as its heating value which depends on the amount of fixed carbon and volatile combustible present in the coal. Besides, the capacity of fuel ignition will be determined by the proportion of high volatile matters in coal whereas the ash content will be a key parameter for designing the furnace or other equipment controlling combustion and pollution [4].

- **Fixed carbon**

After the distillation of volatile matter, the solid fuel remaining in the furnace is known as fixed

carbon. Except for carbon which is its main constituent, it also contains in relatively light proportions some oxygen, hydrogen, sulfur and nitrogen which have not been taken away with gases ejection. Fixed carbon also serves to roughly estimate coal heating value.

- **Ash content**

Coal ash is a complex and variable mixture of minerals that melt over a range of temperatures [5]. Ash in an impurity which doesn't burn away during the whole process. Ash contained in coal varies from 5 to 40%. That value will have many effects on the process such as the reduction of the burning capacity, the decrease in combustion and boiler efficiency and the production of clinker and slag.

- **Volatile matter**

Volatile matters are made up of gases such as methane, hydrogen, carbon monoxide, carbon dioxide, nitrogen and also some other hydrocarbons found in coal. Therefore the presence of volatile matters is closely linked to gaseous fuels contained in the coal. In general, the proportion of volatile matters will vary between 20% and 35%. As for its effects, it can be said that a high content of volatile matters will facilitate coal ignition.

- **Water content**

Since moisture would often replace combustible matters, it will contribute to reduce the heat content found per kg of coal. Typically, water content will be comprised between 0.5 and 10%. During the process of coal transformation, moisture will favor heat loss resulting from evaporation.

- **Sulfur content**

The amount of Sulphur would generally affect the production of clinker and slag. It will also participate in the corrosion of some equipment such as chimney, air heater and economizers. Sulfur also has the capacity of limiting the exit of flue gas temperature.

Some standard values for proximate analysis of some different types of coal are given in the following Table 1.

2.1.2 Ultimate analysis

The ultimate analysis gives the value of some standard chemical elements such as carbon, hydrogen, oxygen or sulfur that are contained in the coal sample. These parameters are important for assessing the amount of air that will be necessary for the combustion process but also to determine the characteristics of combustion gases. This will thus make it easier to estimate the flame temperature and to design the flue duct.

Next is some standard values characterizing the ultimate analysis of various types of biomass.

2.1.3 Coal calorific value, caking index and swelling index

- **Calorific value**

Fuel calorific value, also known as heating value is the amount of heat produced per unit of fuel when coal is heated in the presence of air and at a high temperature. Calorific value can be calculated in two different conditions of constant pressure or constant temperature. We can also distinguish two kind of calorific values namely gross calorific value which corresponds to the heat produced by burning an unit of coal in a water vapor saturated atmosphere at a constant volume; and net calorific value which is determined by subtracting the heat of vaporization of the water vapor from the gross calorific value [7].

Table 1. Standard values of proximate analysis for different types of coal [6]

	Coal rank	M (%)	Ash (%)	VM (%)	FC (%)	HHV (MJ/Kg)
US (Arkansas)	**SA**	2.50	29.30	11.30	56.90	23.56
US (Texas)	**L**	16.36	8.94	37.48	37.22	22.62
India	**B**	9.71	12.88	32.77	44.64	25.74
China	**A**	3.22	13.27	8.46	75.05	29.12
Nigeria	**SB**	6.75	4.51	37.81	50.93	30.14

A: Anthracite, B: Bituminous, L: Lignite, SA: Semi-Anthracite, SB: Sub-Bituminous

Table 2. Standard values of ultimate analysis of different types of coal [6]

	Coal rank	C (%)	H (%)	O (%)	N (%)	S (%)
US (Arkansas)	**SA**	58.30	3.50	3.90	1.20	3.80
US (Texas)	**L**	55.78	5.77	27.40	0.97	1.14
India	**B**	63.35	5.08	16.72	1.40	0.57
China	**A**	73.91	3.38	6.26	1.12	2.06
Nigeria	**SB**	72.09	5.87	15.03	1.85	0.65

A: Anthracite, B: Bituminous, L: Lignite, SA: Semi-Anthracite, SB: Sub-Bituminous

▪ **Caking index**

During the carbonization process, lumps can be produced for certain types of coal. The produced lumps are called cake while caking is a term used to refer to the process of cake formation. The caking index is a parameter that helps to determine coal binding or agglutinating property [8]. The property of caking cannot be found in all kinds of coal and some coals are said to be non-caking. Caking property is essential for industrial purposes when selecting a particular type of coal.

▪ **Swelling index**

Another important property of coal is its swelling index. When coal is heated, a swollen mass is obtained at the end of the process due to the fact that volatile elements have been taken away. This mass is then compared to a standard chart displaying different swollen mass associated to different numbers (0 to 9) and the swelling index will be the number on the chart matching the swollen mass previously obtained [8].

2.2 Properties of Biomass

2.2.1 Proximate analysis

The proximate analysis uses fuel/biomass properties such as moisture, fixed carbon, volatile matter and ash for characterization and classification. These estimations will mostly be done on a mass basis using a specific equipment known as a proximate analyzer [9]. The analyzer proceeds by measuring the mass loss at some specific temperatures. At a temperature between 105ºC and 110ºC (slightly higher than water boiling point), moisture can be determined. This only represents the level of physically bound water inside the biomass [10]. In the case of volatile matters, the process will involve a slow heating rate in a 950ºC inert atmosphere. The value of ash content will be accessed by collecting the material remaining after the loss of volatile matter and burning it in an oxygen-filled environment at above 700ºC. Finally, by subtracting the moisture (M), the ash content and the volatile matter (VM) from 100%, we obtain a value representing the fixed carbon (FC).

$$FC(\%)=100\%-M(\%)-Ash(\%)-VM(\%) \qquad (2.1)$$

Standards values of the proximate analysis of five different types of biomass are given in the following Table 3.

2.2.2 Ultimate analysis

The ultimate analysis is generally conducted to determine various chemical elements such as carbon (C), hydrogen (H), nitrogen (N), sulfur (S), oxygen (O) contained in the biomass. In order to provide reliable data, the ultimate analysis will generally be performed on a dry biomass. If not, water content could be mistaken for additional oxygen and hydrogen [11]. In some cases, the determination of chlorine would be important as it could be a serious source of pollution for processes like gasification, pyrolysis or combustion. This is particularly relevant for straw or other fast-growing biomass materials [12]. Regarding the methods for determining those components, many instruments have been developed and there are cases where C, H, N and S, can altogether be determined at once. Catalytic combustion is used by the majority of systems along with pure oxygen to get nitrogen, carbon dioxide, water and sulfur dioxide from the biomass. Their respective proportion is then calculated by chromatography which relies on the use of either flame ionization detectors or thermal conductivity detectors [13]. The catalytic conversion producing carbon monoxide is often used to access the proportion of oxygen.

Some standards values for ultimate analysis of five different types of biomass are given in the following Table 4.

Table 3. Standard values of proximate analysis of five types of biomass [8]

	M (%)	Ash (%)	VM (%)	FC (%)	HHV (MJ/Kg)
Sarcasa asoca leaf	9.6	5.2	67.9	17.3	20.9
Wheat straw	8.6	7.9	68.4	15.1	17.6
Cotton waste	4.8	3.1	72.1	20.1	16.7
Soybean waste	5.8	4.7	70.5	19.0	18.8
Charcoal	4.6	4.7	18.4	69.1	26.1

Table 4. Standard values of ultimate analysis of five types of biomass [8]

|	C (%)	H (%)	N (%)	S (%)	O (%)
Sarcasa asoca leaf	49.3	6.7	1.2	0.9	42.9
Wheat straw	40.6	6.0	0.2	0.9	53.2
Cotton waste	50.5	7.0	1.2	0.8	22.5
Soybean waste	43.8	6.3	1.4	0.8	48.5
Charcoal	72.2	2.9	1.3	0.7	23.6

3. DEVOLATIZATION PROCESS IN COAL AND BIOMASS GASIFICATION

3.1 Devolatization Process of Coal

When coal enters a gasifier, it is first subjected to a drying process.

$$H_2O(l) \rightarrow H_2O(g) \qquad \Delta H°_{vap} = 44kJ/mol \text{ (3.1)}$$

A similar format to what will be used for chemical reactions will be used to write this transition phase. Water vaporization can have a dominant effect on the overall gasification thermodynamics, especially when a high moisture, low-grade coal is processed, or when the coal is fed to the reactor as a coal-water slurry. Water vaporization will have a limited impact in the case of bituminous coals or when using a dry feed gasifier. Coal pyrolysis would also produce coke and volatiles:

$$Coal \rightarrow coke(s) + volatiles(g) \qquad \Delta H°_{rxn} = positive, variable \text{ (3.2)}$$
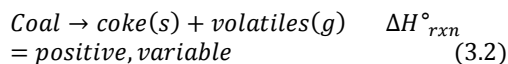
The above reaction is a simplified way of describing a complex and variable set of reactions. When temperatures reach and surpass 360°C, carbon-carbon bonds, or bonds between carbon and oxygen, nitrogen, or sulfur, in the backbone of organic compounds break. At the very beginning of the reaction, unstable molecular fragments are formed. Those fragments may further undergo pyrolysis, or react to form relatively stable compounds. When cooled at ambient temperature, pyrolysis will lead to gaseous or liquid products in the form of coal tar or solid products in the form of soot.

Due to coal complexity, and the interdependence between pyrolysis yields and processing conditions, an empirical approach for modeling reaction (3.2) is often taken. Some scholars have developed a chemical percolation model for devolatization (CPD) that uses structural data from coal and ^{13}C NMR analysis to predict pyrolysis yields [14]. Fig. 1 illustrates concepts used to quantify coal structure. Coal structure includes aromatic clusters which are made up of a single aromatic ring or multiple fused aromatic rings. The clusters are connected by aliphatic bridges. Side chains are aliphatic groups that may be bridged or not. A loop is a situation where a side chain is connected to the same aromatic group in more than one location. The parent coal is assumed to have an infinite molecular weight.

Pyrolysis begins when a bridge breaks down, creating a nonvolatile, finite molecular weight fragment known as a metaplast. During pyrolysis, coal typically undergoes a semi-liquid intermediate phase; leading to the formation of solid chars and volatiles. From the electron microscope images of rapidly heated char particles, it can be seen that the char particle is often a hollow sphere, which is formed by an expanding gas bubble in a semi-liquid matrix [16].

The metaplast is subjected to one of two antagonist reactions. The first reaction consists of splitting it into two smaller fragments whereas the second reaction enables the formation of char along with two smaller fragments. Raoult's law and a correlation between pure component vapor pressure and molecular weight are both used to describe the product volatility.
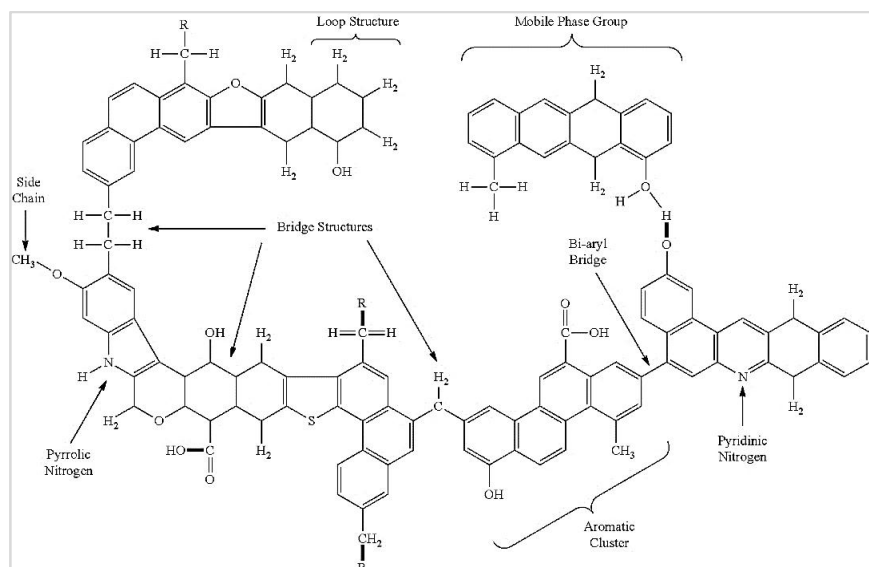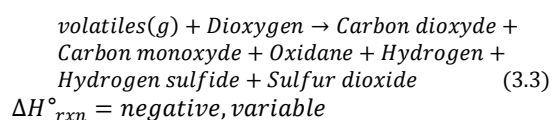
Fig. 1. Coal structure information measured using 13C NMR and then used to predict volatiles yield using the CPD model [15]

A major hypothesis in the chemical percolation model for devolatization is that the coal type does not influence reaction rate constants. Instead, variation in pyrolysis yields is due to structural differences, which can be measured using ^{13}C NMR. The model has been successfully applied to a wide range of coals and can be said to be satisfactory considering the chemical variability and complexity of coal. Since ^{13}C NMR data for coal are not widely available, correlation approaches have been used for determining the CPD structural parameters using more commonly obtained coal assay data [17].

Majority of the volatile fraction will undergo reactions to form soot [18].

If there are not enough spaces between feed points for coal and oxygen (or air) in the gasifier, then the volatiles may burn to produce combustion and partial-combustion products:

$$volatiles(g) + Dioxygen \rightarrow Carbon\ dioxyde + Carbon\ monoxyde + Oxidane + Hydrogen + Hydrogen\ sulfide + Sulfur\ dioxide \quad (3.3)$$
$$\Delta H^{\circ}_{rxn} = negative, variable$$

In moving bed gasifiers with relatively low temperatures, the pyrolysis process takes place between 700 to 800ºC. Coal and oxygen move through the reactor in a countercurrent way, and most of the oxygen is depleted before it reaches the pyrolysis section of the gasifier. Consequently, this type of gasifier generates large quantities of coal tar. Fluidized bed gasifiers, work at about 1000ºC, and the feed points are often similar for coal and oxygen. These gasifiers produce little tar, but a considerable amount of methane. The experimental temperature does not enable higher molecular weight volatile products to be stable which are therefore not produced. Entrained flow gasifiers, characterized by high temperatures between 1400ºC and 1500ºC, produce no coal tar and little amount of methane [19].

3.2 Devolatization Process of Biomass

The process of pyrolysis consists of heating biomass or other feed in the absence of air or oxygen, at a specified rate and at a maximum temperature, keeping it in that condition for a specific duration. The heating rate and the pyrolysis temperature are the main influencing factors concerning the obtained products. Condensable gases and solid char constitute the pyrolysis first products.

The condensable gases may break down further into non-condensable gases (CO, CO2, H2, and CH4), liquid, and char (Fig. 2). This decomposition occurs partly through gas-phase homogeneous reactions and also through gas-solid-phase heterogeneous thermal reactions. In gas-phase reactions, the condensable vapor is broken down into smaller molecules of non-condensable permanent gases such as CO and CO2 [12].
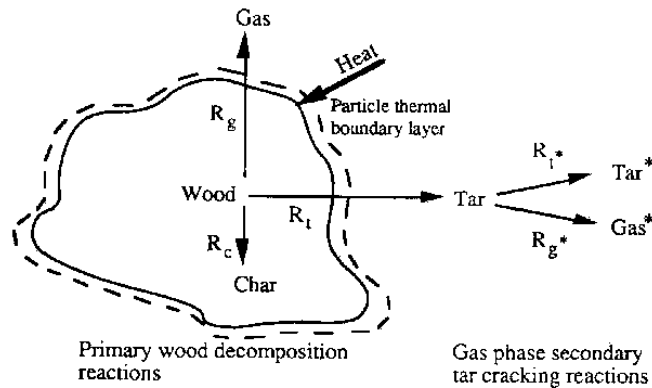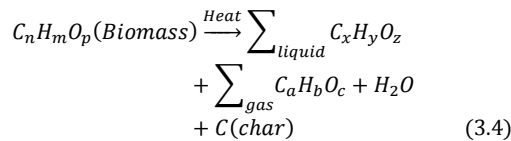
Fig. 2. Pyrolysis in a biomass particle

The pyrolysis process may be represented by a generic reaction as follows:

$$C_nH_mO_p(Biomass) \xrightarrow{Heat} \sum_{liquid} C_xH_yO_z$$
$$+ \sum_{gas} C_aH_bO_c + H_2O$$
$$+ C(char) \qquad (3.4)$$

One important pre-step in any gasifier is pyrolysis. It is relatively fast, especially in reactors with rapid mixing.

We can classify pyrolysis products into three major categories:

- **Solid:** Char is the solid yield of pyrolysis. It primarily contains carbon ($\square$85%), but oxygen and hydrogen can also be found in little quantity. Unlike fossil fuel, biomass contains very little inorganic ash. The lower heating value (LHV) of biomass char is about 32 MJ/kg, which is substantially higher than that of the parent biomass or its liquid product [20].
- **Liquid:** The liquid yield, known as tar, bio-oil, or bio-crude, is a very dark tarry fluid containing up to 20% water. It is principally made up of homologous phenolic compounds. Bio-oil is a mixture of complex hydrocarbons with large amounts of oxygen and water. While the parent biomass has an LVH in the range of 19.5 to 21MJ/kg dry basis, its liquid yield has a lower LVH in the range of 13 to 18MJ/kg wet basis [21].
- **Gas:** Primary decomposition of biomass yields two products namely condensable gases (vapor) and non-condensable gases (primary gas). In the case of vapors with heavier molecules, they will condense

upon cooling and end up as liquid yield. On the other hand, lower-molecular-weight gases like carbon dioxide, carbon monoxide, methane, ethane, and ethylene are part of the non-condensable gas mixture.

Based on the heating rate, pyrolysis can generally be classified as fast or slow. It will be said to be slow if the time needed to heat the fuel to the pyrolysis temperature ($t_{heating}$) is much longer than the typical pyrolysis reaction time (t_r) and fast in the opposite case. That is:

- Slow pyrolysis: $t_{heating} \gg t_r$
- Fast pyrolysis: $t_{heating} \ll t_r$

These criteria may be expressed in terms of heating rate as well, assuming a simple linear heating rate ($T_{pyr}/t_{heating}, K/s$). The characteristic reaction time, t_r , for a single reaction is taken as the reciprocal of the rate constant, k, evaluated at the pyrolysis temperature [22].

Products of pyrolysis not only depend on the model of pyrolyzer, but also on physical and chemical characteristics of the biomass, and other important factors such as the heating rate, the pyrolysis temperature and the amount of time spent in the reaction zone.

Besides these, the tar and the yields of other products depend on pressure, ambient gas composition, and the presence of mineral catalysts [23]. Yields of solid, liquid and gaseous products of pyrolysis can possibly be modified by changing the final temperature and the heating rate. Fast heating leads to higher volatiles and more reactive char compared to a slower heating
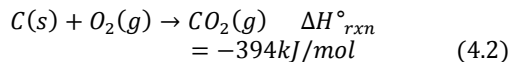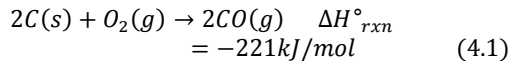
process; slower heating rate and longer processing time result in the production of a secondary char from the reaction between the primary char and the volatiles.

4. CHAR GASIFICATION REACTIONS

4.1 Coal char Gasification Reactions

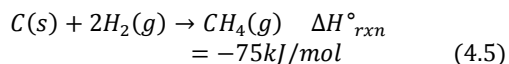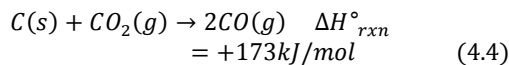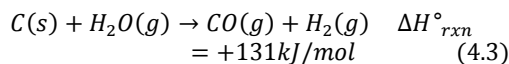#### 4.1.1 Reactions with oxygen

Oxygen enters the gasifier either as a nearly pure stream of oxygen or as air. The reaction between oxygen and the char produces carbon monoxide or carbon dioxide:

$$2C(s) + O_2(g) \rightarrow 2CO(g) \quad \Delta H°_{rxn} = -221 kJ/mol \quad (4.1)$$

$$C(s) + O_2(g) \rightarrow CO_2(g) \quad \Delta H°_{rxn} = -394 kJ/mol \quad (4.2)$$

The overall heat of reaction can be reduced through above reactions. For example, Reaction 4.1 produces two moles of carbon monoxide, and the heat of reaction is twice the heat of formation of carbon monoxide. On the other hand, reaction 4.2 produces one mole of carbon dioxide, so the heat of reaction is the same as the heat of formation for carbon dioxide.

4.1.2 Other char reactions

In addition to the combustion of char, the following reactions can also be considered:

$$C(s) + H_2O(g) \rightarrow CO(g) + H_2(g) \quad \Delta H°_{rxn} = +131 kJ/mol \quad (4.3)$$

$$C(s) + CO_2(g) \rightarrow 2CO(g) \quad \Delta H°_{rxn} = +173 kJ/mol \quad (4.4)$$

$$C(s) + 2H_2(g) \rightarrow CH_4(g) \quad \Delta H°_{rxn} = -75 kJ/mol \quad (4.5)$$

Reaction 4.6 is also known as the steam gasification reaction. Reaction 4.7 is the CO_2 gasification reaction, also known as the Boudouard reaction. Reaction 4.8 is the methanation reaction [19].

4.1.3 Additional gas phase reactions

In order to adjust carbon monoxide and hydrogen ratios, the water gas shift reaction is often used in a catalytic reactor downstream of the gasifier. In the gasifier, this reaction occurs immediately because of the reaction's high temperature:

$$CO(g) + H_2O(g) \leftrightarrow CO_2(g) + H_2(g) \quad \Delta H°_{rxn} = -41 kJ/mol \quad (4.6)$$
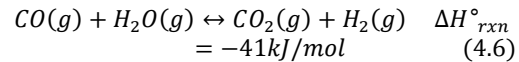
The production of syngas from natural gas is achieved through the use of the steam methane reforming reaction in catalytic reactors. High temperature would also favor the instantaneous occurrence of this reaction in the gasifier:

$$CH_4(g) + H_2O(g) \leftrightarrow CO(g) + 3H_2(g) \quad \Delta H°_{rxn} = 206 kJ/mol \quad (4.7)$$
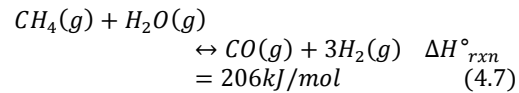
4.1.4 Balance coal, oxygen and water feeds

Reactions, 4.2, 4.3, and 4.4, are very exothermic and release a great amount of heat, but the product gasses have no further combustion value. The steam and CO_2 gasification reactions (4.6 and 4.7), are endothermic; which means that the product gasses have a greater heating value than the reactants [19].

By modifying the balance between oxygen and water, the temperature can be controlled. Reactions with oxygen would all produce heat and so oxygen would tend to increase the gasifier temperature. The steam gasification reaction, 4.6, also leads to gas formation. Steam gasification is endothermic and therefore reduces the gasifier temperature. To optimize gasifier operation, it is important to find the correct ratios of $(O_2 + H_2O)$/coal and O_2/H_2O.

Steam can be replaced by carbon dioxide, but the CO_2 gasification reaction (4.7), is slower than the steam gasification reaction (4.6). Besides, the higher H_2/CO ratio produced by the steam gasification reaction is advantageous for most syngas applications.
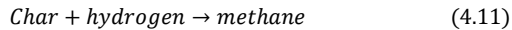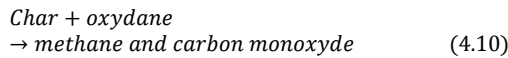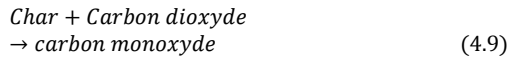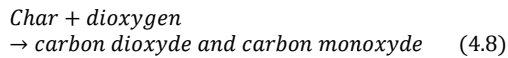
4.1.5 Air versus oxygen feed

The most affordable source of oxygen for gasification is compressed air. However, that method would favor a large quantity of nitrogen along with argon to enter the gasifier. Moisture-free air is about 20.95% oxygen, 78.08% nitrogen, 0.93% argon, and 0.40% other gasses. Reactive gasses are diluted by nitrogen and argon, which are mostly inert. Additional oxygen is required to raise these inert molecules to the reaction temperature [19].

4.2 Biomass Char Gasification Reactions

On a general basis, biomass char is more porous and reactive than coke. Its porosity is in the range of 40 to 50% while that of coal char varies between 2 and 18%. Biomass char has much larger pores (20-30 micron) than coal char (Σ5 angstrom) [24]; thus, there is a large difference between biomass reaction behavior and that of chars derived from coal, lignite, or peat. For example, the reactivity of peat char decreases with conversion or time, unlike biomass char which reactivity increases with conversion (Fig 3). This reversed trend can be caused by the increased catalytic activity of the biomass char's alkali metal constituents [25].

Gasification of biomass char includes several reactions involving char and gasifying agents. What follows is some of those reactions with carbon dioxide, hydrogen, stem, and methane.

$$Char + dioxygen \\ \rightarrow carbon\ dioxyde\ and\ carbon\ monoxyde \quad (4.8)$$

$$Char + Carbon\ dioxyde \\ \rightarrow carbon\ monoxyde \quad (4.9)$$

$$Char + oxydane \\ \rightarrow methane\ and\ carbon\ monoxyde \quad (4.10)$$

$$Char + hydrogen \rightarrow methane \quad (4.11)$$

Equations 4.12 to 4.15 present reactions between some gasifying agents including oxygen, carbon dioxide, steam and solid carbon to produce carbon monoxide.

Gasification reactions are in most cases endothermic, but some can also be exothermic. For example, the reactions of carbon with oxygen and hydrogen are endothermic.

4.2.1 Speed of char reactions

The gasification rate of char (mainly made up of carbon) relies primarily on char reactivity and the sensitivity level of the gasifying medium. For gasification, the most active agent is oxygen, followed by steam and carbon dioxide. The rate of the char-oxygen reaction ($C + \frac{1}{2}O_2 \rightarrow CO$) is one of the fastest. During that reaction, oxygen is consumed within a very short period of time, leaving almost no free oxygen for any other reactions.

The rate of char-stream reaction ($C + H_2O \rightarrow CO + H_2$) is three to five times slower than that of the char-oxygen reaction. The Boudouard, or char-carbon dioxide reaction ($C + CO_2 \leftrightarrow 2CO$) is six to seven orders of magnitude slower [26]. The rate of the water-gas or water-steam gasification reaction ($C + H_2O \leftrightarrow CO + H_2$) is about two or five times faster than that of the Boudouard reaction [27].
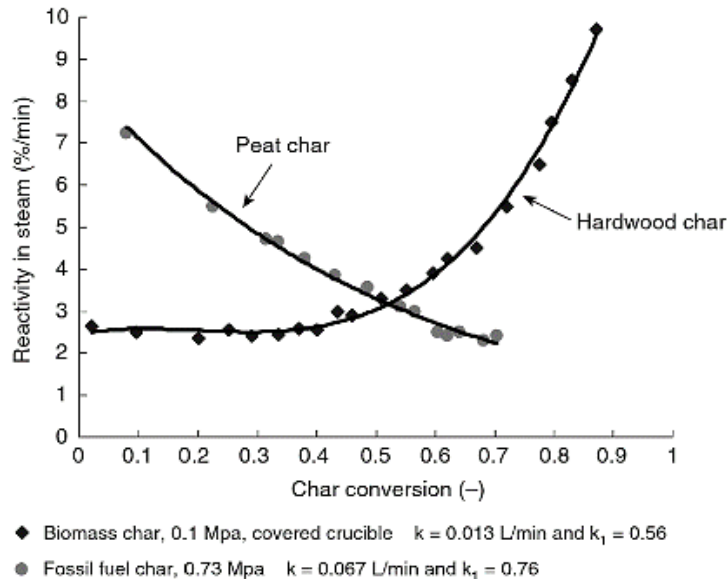

◆ Biomass char, 0.1 Mpa, covered crucible   k = 0.013 L/min and k_1 = 0.56

● Fossil fuel char, 0.73 Mpa   k = 0.067 L/min and k_1 = 0.76

Fig. 3. Peat char and Hardwood char behavior with the variation of reactivity and char conversion [12]

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The char-hydrogen reaction that forms methane ($C + 2H_2 \rightarrow CH_4$) is the slowest of all. The relative rate of the four reactions, at 800°C temperature and 10K Pa pressure, is estimated as 10^5 for oxygen, 10^3 for steam, 10^1 for carbon dioxide, and 3×10^{-3} for hydrogen [28].

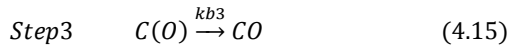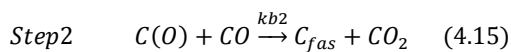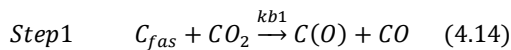$$R_{C+O_2} \gg R_{C+H_2O} > R_{C+CO_2} \gg R_{C+H_2} \quad (4.12)$$

The reaction between steam and carbon can produce CO and H_2. In certain cases methane and carbon dioxide can also be produced from the steam and carbon reactions.

4.2.2 Boudouard reaction model

The name Boudouard reaction is often used to refer to the gasification of char in carbon dioxide.

$$Carbon + Carbon\ dioxyde \\ \leftrightarrow 2Carbon\ monoxyde \quad (4.13)$$
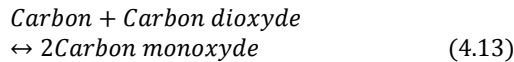
The Boudouard reaction can be described through different steps. In the first step, CO_2 dissociates from a carbon-free active site (C_{fas}), releasing carbon monoxide which generates a carbon-oxygen surface complex $C(O)$. This reaction can also occur in the reverse way, forming a carbon active site and CO_2 in the second stage. During the third stage, the carbon-oxygen complex produces one molecule of CO[27].

$$Step1 \qquad C_{fas} + CO_2 \xrightarrow{kb1} C(O) + CO \quad (4.14)$$

$$Step2 \qquad C(O) + CO \xrightarrow{kb2} C_{fas} + CO_2 \quad (4.15)$$

$$Step3 \qquad C(O) \xrightarrow{kb3} CO \quad (4.15)$$

Where k_i is the rate of the ith reaction.

The rate of the char gasification reaction in CO_2 is very low and below 1000K.
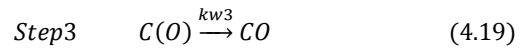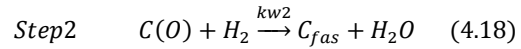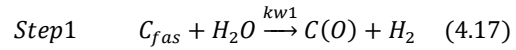
4.2.3 Water-gas reaction model

One of the most important gasification reaction might be the gasification of char in steam, known as the water-gas reaction.

$$Carbon + Oxydane \\ \leftrightarrow Carbon\ monoxyde \\ + Hydrogen \quad (4.16)$$
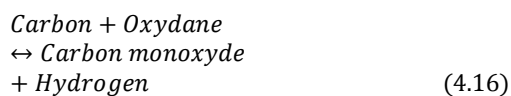
The first step forms a surface oxide complex of carbon CO, while in the second and third step,

the surface oxide complex produces a new free active site

$$Step1 \qquad C_{fas} + H_2O \xrightarrow{kw1} C(O) + H_2 \quad (4.17)$$

$$Step2 \qquad C(O) + H_2 \xrightarrow{kw2} C_{fas} + H_2O \quad (4.18)$$

$$Step3 \qquad C(O) \xrightarrow{kw3} CO \quad (4.19)$$

The possibility of hydrogen inhibition by $C(H)$ and $C(H)_2$ is included in some models as here:

$$C_{fas} + H_2 \leftrightarrow C(H)_2 \quad (4.20)$$

$$C_{fas} + 0.5H_2 \leftrightarrow C(H) \quad (4.21)$$

The presence of hydrogen has a high inhibiting effect on the char gasification rate in H_2O. For example, 30% hydrogen in the gasification atmosphere can reduce the gasification rate by a factor of 15 [29]. So an effective method of accelerating the water-gas reaction is to keep removing hydrogen from the reaction zone.

4.2.4 Shift reaction model

The shift reaction is an important gas-phase reaction. It increases the quantity of hydrogen content of the gasification product by reducing carbon monoxide. This reaction is also called the "water-gas shift reaction", though there is a difference with the water-gas reaction ($C + H_2O \leftrightarrow CO + H_2$).

$$Carbon\ monoxyde + Oxydane \\ \leftrightarrow Carbon\ dioxyde \\ + Hydrogen \\ - 41kJ/mol \quad (4.22)$$
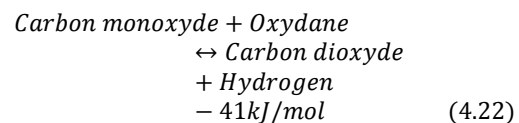
This is a preliminary phase of syngas production in the downstream of a gasifier, where the hydrogen and carbon monoxide ratio in the product gas is critical.

The shift reaction releases a small amount of heat, and its equilibrium yield decreases slightly with temperature. Depending on temperature, it may end up either as product or reactant. However, it is not influenced by pressure [30].

At temperatures higher than 1000ºC the shift reaction (4.27) rapidly reaches equilibrium, but at lower temperatures, heterogeneous catalysts would be needed. From Fig. 4 we notice that this reaction has a higher equilibrium constant at a
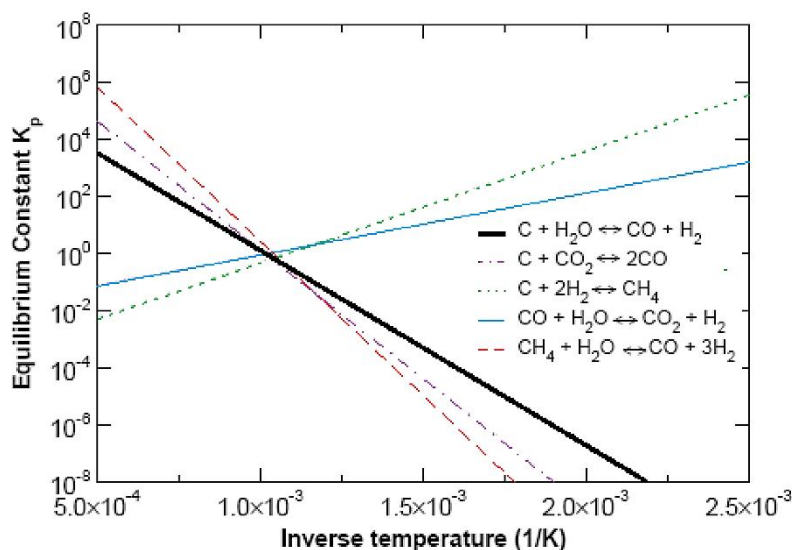
Fig. 4. Equilibrium constants for selected gasification reactions [31]

lower temperature, which implies a higher yield of H_2 at a lower temperature [29]. As temperature increases, the yield decreases but the reaction rate increases. Optimum yield is obtain at about 225℃ [31].

4.2.5 Hydrogasification reaction model

This reaction consists of the gasification of char in an environment containing hydrogen, leading to the production of methane.

$$Carbon + 2Hydrogen \rightarrow Methane \qquad (4.23)$$
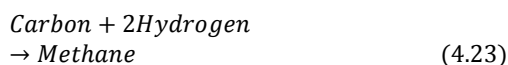
This reaction has a much slower speed than others. It can only be useful when the production of the synthetic natural gas (SNG) is needed.

5. SIMILITUDE AND CONTRAST

5.1 The Atomic Ratio

When the comparison is made based on the atomic ratio, it is easier to understand the heating value of a fuel. For example, the higher heating value (HHV) of biomass correlates well with the oxygen-to-carbon (O/C) ratio. The HHV will decrease from 38 to about 15 MJ/kg when the O/C ratio increases from 0.1 to 0.7 [12]. With the increase in hydrogen-to-carbon (H/C) ratio, the effective heating value of the fuel is reduced. The atomic ratio depends on hydrogen, oxygen, and carbon content of the fuel.

Fig. 5 shows the atomic ratios (H/C) and (O/C) on a dry ash free basis for all fuels, from carbon-rich anthracite to carbon-deficient woody biomass. From this plot, known as the "Van Krevelen diagram", the biomass has much higher ratios of H/C and O/C than fossil fuel. For some specific types of biomass, a linear function can be drawn between H/C and O/C ratios.

It is possible to have very low heating values with fresh plants biomass like leaves because of their high H/C and O/C ratios. The atomic ratio of a fuel will decrease when its geological age increases, which means that as the fuel age increases, its energy content increases too.

As an example, a fossil fuel geologically formed over many thousands of years like Anthracite has a very high heating value. Its lower H/C favors heat production, but CO_2 emission from its combustion is high.

Compared to other hydrocarbon fuels, biomass has the highest proportion of oxygen. Unfortunately, oxygen does not make any useful contribution to heating value and increases the difficulty to transform the biomass into liquid fuels. The high oxygen and hydrogen content of biomass results in highly volatile and liquid yields, respectively [12]. High oxygen will consume a part of the hydrogen in the biomass leading to the production of less beneficial water, and thus a high H/C content does not translate into high gas yield.
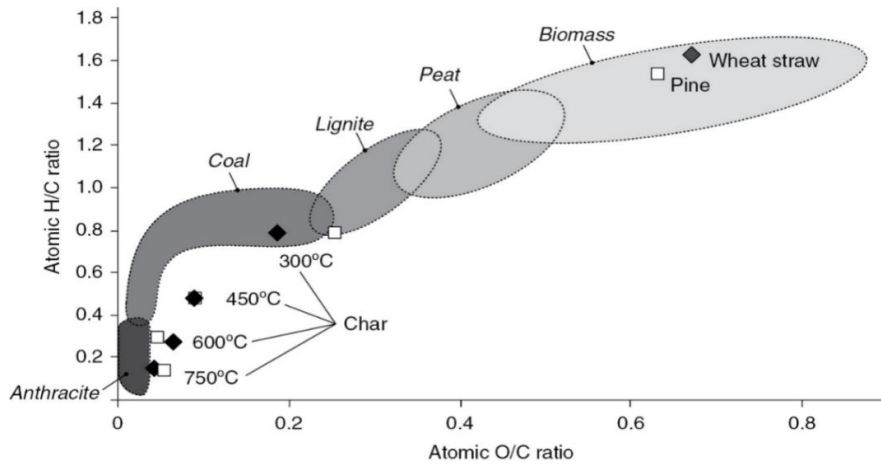
11

Fig. 5. Classification of solid fuels by their hydrogen/carbon and oxygen/carbon ratios [32]

5.2 Carbon Conversion

Carbon conversion during gasification will vary according to the temperature and CO_2 concentration in the feed gas. The observations made from Fig. 6 are as follows:

- Carbon conversion is more dependent on temperature variation than CO_2 concentration. As observed in the case of Coal and Biomass, the increase in CO_2 concentration corresponds to an increase in conversion rate of 6% at lower temperatures. On the other hand, the temperature rise results in the increase of 8% in the case of coal and 10% in the case of biomass [33]. The conversion values for

biomass is significantly higher compared to coal at 1200ºC, based on solids basis calculations.

- The CO_2 requirement for gasification is an important parameter that should be obtained from lab-scale gasification studies. Considering equilibrium conditions, approximately 2 LCO_2/g carbon fed is required to provide pure CO_2 feed under the reaction conditions. As seen from Fig. 6, except for the case "10% CO_2" concentration, the CO_2 is fed in excess. The excess CO_2 released from downstream applications can either be re-injected into the feed stream or be used for chemical synthesis.
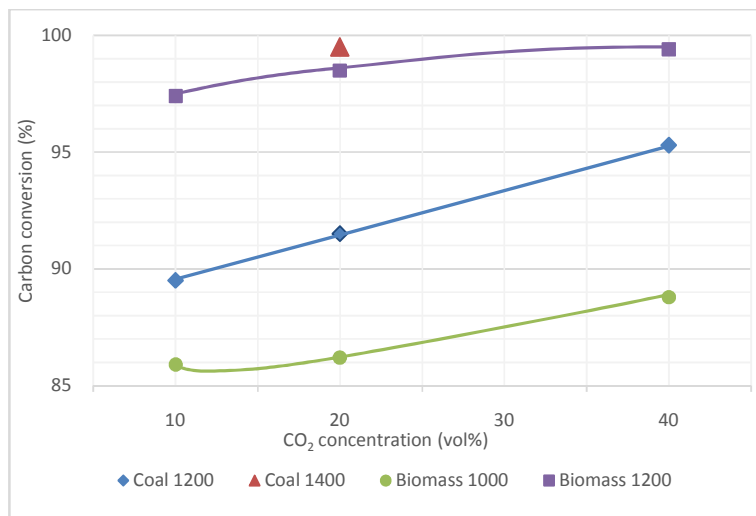

Fig. 6. Carbon conversion on solids basis obtained for coal (Victorian brown coal) and biomass (pine bark) at different temperatures represented as a function of CO2 concentration in the feed gas. Figures in parenthesis indicate the amount of CO2 in the feed gas in L/min

5.3 Syngas Composition

The influence of temperature and CO_2 concentration on syngas composition (N_2 free basis) from Coal and Biomass gasification is presented in Fig. 7. We can observe that as the temperature increases, the CO concentration increases too but decreases along with the decrease of the CO_2 concentration in the feed. On the other hand, the production of H_2 decreases with both temperature and CO_2 concentration within the feed. With the increase in CO_2 percentage in the feed stream, the proportion of unreacted feed CO_2 increases too, thus leading to a corresponding decrease in CO and H_2 concentration [34].

From Fig. 7, the comparison between the fuels regarding the overall gas composition indicates that CO/H_2 ratio is lower for the coal sample (13:1) in comparison to the biomass (18:1) at 1200ºC 20% CO_2. The CO/H_2 ratio for coal has a range of variation between 5:1 and 21:1, whereas for biomass, the ratio varies between 4:1 and 38:1. The lower syngas ratios are obtained at the lowest temperature i.e., 1000ºC for biomass and 1200ºC for coal for 10% CO_2 in N_2 as the gasifying agent. However, these conditions do not result in perfect conversion ratio. Therefore, the realistic CO/H_2 ratio to be expected from CO_2 gasification of these fuels under entrained flow conditions is in the range of 20:1–30:1. Syngas generated from CO_2 gasification is expected to be rich in CO. If the gasification is used for chemical synthesis, this ratio may not be suitable. Therefore, the syngas ratio will have to be adjusted using the water gas shift reaction to lower the syngas ratio typically down to 2:1 (for methanol) [35].

On the other hand, if the gasification is conducted for power generation purposes, then the heating values for the generated syngas are important parameters to consider. On a general basis, heating values (HHV) for the coal (3.58–8.48 MJ/m3) are higher than those for biomass (0.89–4.24 MJ/m3). Considering the fact that the exit gas is diluted with CO_2, it is very likely to still have low heating values. Therefore, even for power generation, the water gas shift reaction has to be carried out to improve the heating value of syngas [33].

5.4 Char Characterization

Fig. 8 is a distribution diagram of particles size for various chars of coal and biomass samples.

The comparison has been made in terms of d (0.1), d (0.5) and d(0.9) which are defined as the particle size at 10, 50 and 90% of the total volume distribution. For coal samples, chars have the similar d (0.9) values of around 150 µm compared to the parent coal. Furthermore, the same trend is observed for d(0.5) values that are consistently in the mean particle size range of 90–106 µm. However, in the case of d (0.1), there is a large increase in the size of chars compared to the parent coal. From Fig. 8 only, it is difficult to define a rigorous trend regarding particle size evolution for coal chars with both CO_2 concentration and temperature. However, we can notice that d(0.1) values decreased by 10–15 µm when the temperature increased from 1200 to 1400ºC at 20% CO2. On the other hand, in the case of biomass, there is a lot of similarity with the observations made on coal; however, a large variance is observed in d (0.9) values. The large variance could be explained by the agglomeration of char particle at higher temperatures resulting in particles with much larger sizes [33]. Another observation specific to biomass is that d (0.1) values at a higher temperature are lesser than for raw biomass.

The particle sizes of chars are mainly dependent on three factors namely particle agglomeration, particle swelling, and particle fragmentation. Agglomeration (or coalescence) of the char particles happens due to the melting mineral constituents. The ash composition and fusion are very important when it comes to defining the agglomeration between char particles. Agglomeration has been observed for both coal and biomass in fluidized bed gasifiers which result in particle de-fluidization problems [36].

Looking at the char morphology, it can be said that in the parent fuel, the biomass sample had a fibrous structure whereas the coal was characterized by a dense smooth surface. A clear difference was observed in the production of char particles from these fuels. Carbon-rich char particles were distorted in appearance compared to the mineral-rich ones which appeared to maintain their original shape or take a spherical shape due to mineral melting [33]. At higher temperatures, mineral constituents in the chars were disposed on the surface of the particle.

5.5 Pollutant Gas Emission

It has been shown that temperature and CO_2 concentration have a great influence on H_2S,
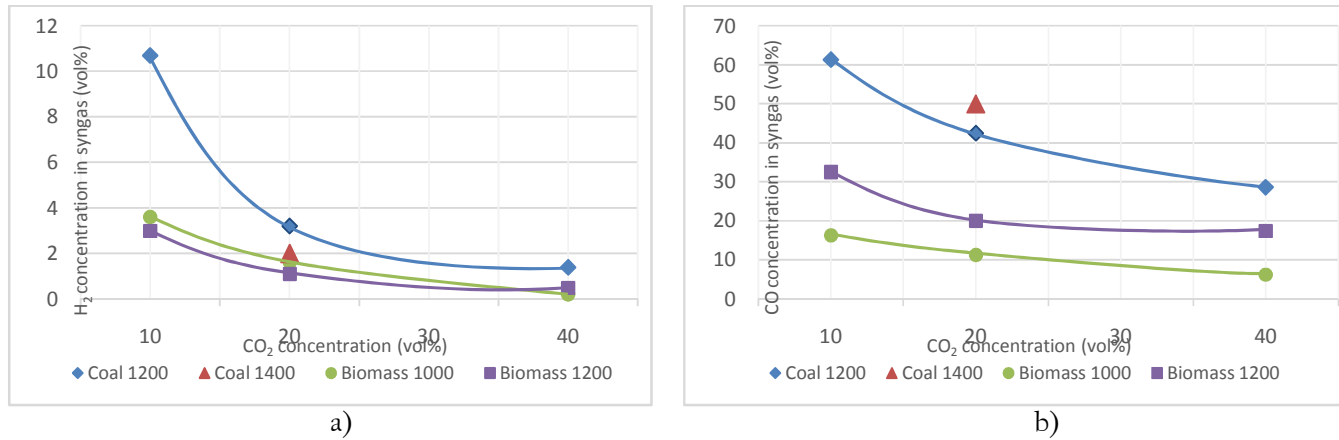
a) b)

Fig. 7. Syngas composition (N2 free basis) [a- for H$_2$; b- for CO] obtained for coal (Victorian brown coal) and biomass (pine bark) at different temperatures represented as a function of CO2 concentration in the feed gas
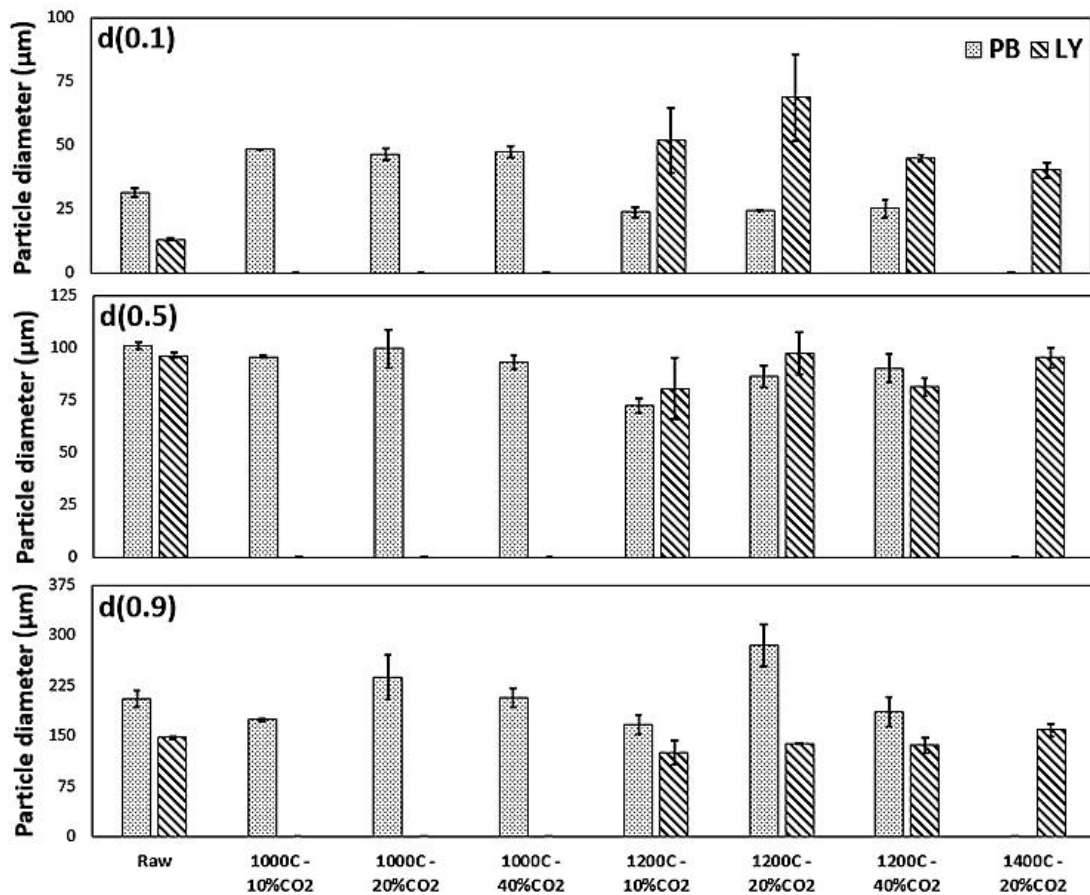
Fig. 8. Particle size distribution for the raw sample and the chars of (a) coal (Victorian brown coal) (b) biomass (pine bark) [33]

HCN, and NH_3 release for coal and biomass feedstocks. The concentration of these pollutants has been evaluated in terms of ppmv range for both feedstocks. However, there is a significant difference between the concentration of these pollutants released during the gasification of coal and biomass. Considering H_2S, the coal sample releases significantly higher H_2S compared to biomass. The concentration range for coal is between 40 and 80 ppmv; whereas for biomass, the concentration is 2.5 ppmv which is relatively negligible [37]. The result is worth analyzing since both coal and biomass have very low sulfur contents around 0.12% and 0.13 % of the sample total weight. Therefore, the large difference in H_2S release profiles can be explained by the nature of the sulfur in the organic matrix. In coal, about 60% of the sulfur is known to be organically bonded resulting in an H_2S release. In biomass, on the other hand, most of the sulfur is inorganically bonded. Sulfur is also produced mainly through thermal cracking

and hydrogenation of the sulfur-containing molecules [37]. Besides, S atoms inside the mineral matter can contribute to the H_2S release. The H_2S release in coal may be due to two factors: the thermal decomposition of organic sulfur at higher temperatures [38] and the consumption of H_2S to form COS as illustrated by the following equation

$$H_2S_{(g)} + CO_{2_{(g)}} \leftrightarrow COS_{(g)} + H_2O_{(g)} \qquad (5.1)$$
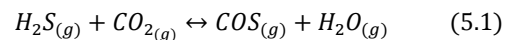
The first factor is an explanation to the increase in H_2S release with temperature and the second explains the decrease in H_2S when CO_2 increases in the feed gas.

6. TECHNOLOGICAL PROGRESS AND FUTURE DEVELOPMENT

One of the major challenges of the 21st century is to propose new techniques in order to satisfy the global energy demand while minimizing the

impact on the environment. Gasification, as a reliable and flexible technology, will be an important asset for this new energetic revolution, as it proposes various technologies to boost clean energy production [39].

6.1 Progress in Coal Gasification Technologies

Coal as the most used primary fossil fuel around the world for energy production is generally at the origin of CO_2, SO_x, NO_x emissions and other noxious compounds.

In recent years, some innovations have been made to tackle that problem and to further increase the gross thermal power generation efficiency to a rate as high as 89% (Fig. 10) [40]. Some of those promising research include the following technologies:

- Hydrogen-from-coal process (HYCOL): This gasification technology enables pulverized coal to be gasified in an entrained bed gasifier using oxygen as the gasifying agent at a very high temperature (ranging from 1500ºC to 1800ºC) and pressure (generally 3Mpa). At the end of the gasification process, two gases will mainly be produced namely hydrogen (31%) and carbon monoxide (61%) and the reaction will perform a rate of carbon conversion of 98%and will reach a cold gas efficiency of 78% [40]. There are also some major features characterizing this technology; the first is this technology uses the method of two stages swirling-entrained-bed in one chamber, the second is the ash contained in the slag can be ejected via a slag hole created at the middle of the gasifier and the third is a cyclone can be used to separate unreacted char which will then be recycled at high temperature and high pressure.
- CO_2-recovery-type IGCC (Integrated Gasification Combined Cycle) system: In this technology, a portion of CO_2 in the exhausted gas is recycled and used as the gasifying agent with the cryogenic air separation unit producing O_2 [41]. Some characteristics of this system are that oxygen is used in the gasifier to significantly reduce the energy needed for the separation of CO_2 from the exhausted gas; the CO_2 recycled into the gasifier can

enhance coal gasification as shown in the equation $C + CO_2 \rightarrow 2CO$, and thus improve cold gas efficiency. In the case of CO_2 recovery, the gross power efficiency can reach a ratio of 42%.

- Advanced IGCC/IGFC (Integrated Gasification Fuel Cell) with exergy recovery technology: Following the exergy recuperation theory, the concepts of Advanced IGCC (A-IGCC) and Advanced IGFC (A-IGFC) were developed, in which high-temperature gasifiers are replaced by a steam reforming gasifier, and the energy for steam gasification is obtained through the recycling of exhausted heat from gas turbine or solid oxide fuel cells (SOFCs). Thus, the power generation efficiency is higher than that of conventional IGCC and IGFC by about 10%. To realize that, a high-density triple-bed combined circulating fluidized bed (TBCFB) gasifier system is used, which includes a downer pyrolyzer, a bubbling fluidized bed (BFB) gasifier and a riser combustor [42].
- Super IGFC (S-IGFC): This technology was recently introduced as a coal gasification method with power generation efficiency reaching 89% [40]. The main constituents of S-IGFC are a steam gasifier and a SOFC (Solid Oxide Fuel Cell), in which the heat and steam generated in SOFC converge to the steam gasifier, and no gas or steam turbine is needed for this system. In particular, compared to other conventional gasification techniques, S-IGFC will be operated with a small amount of oxygen, at a relatively low temperature and the produced syngas will be rich in methane (15% to 30%) and also contain hydrogen and carbon monoxide. Besides, the high concentration in methane contained in the produced syngas can be processed and contribute in the cooling module, thereby helping to reduce the need for energy supply from an external source.

6.2 Progress in Biomass Gasification

Biomass as a renewable energy source has an enormous potential for heat, power, and biofuels production. Consequently, it is one of the most explored research areas in the field of sustainable energy.
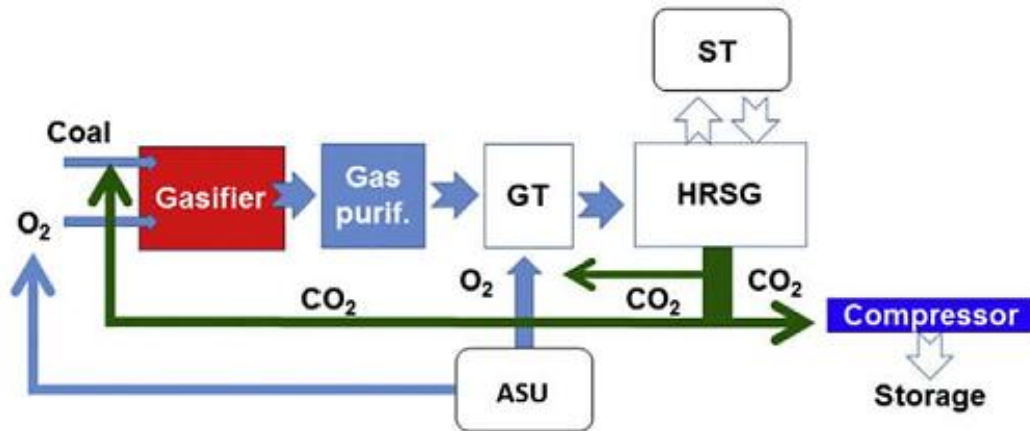
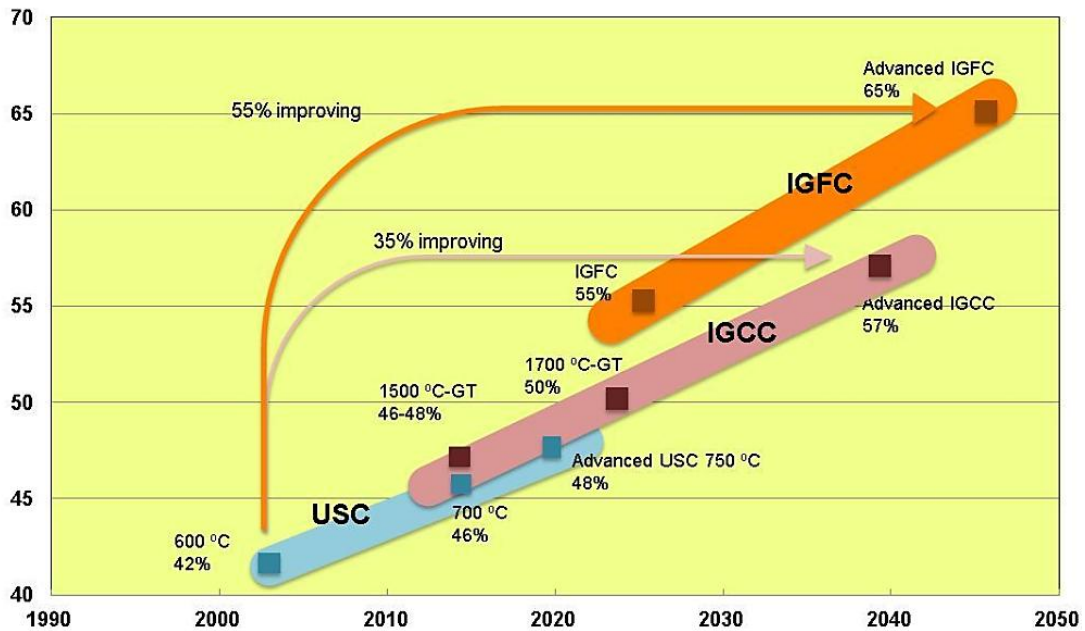**Fig. 9. Proposed CO2-recycle-type IGCC system [40]**

Fig. 10. Net generation efficiency (coal gasification), HHV (%) [41]

Many innovative, cost-effective and efficient gasification techniques are being developed in that sense.

- Supercritical Water gasification: It is a promising concept which will help to convert wet biomass with high moisture into syngas. As there would be no need to resort to energy-consuming drying of the wet biomass, thus energy efficiency would be enhanced. Wet biomass with a moisture of more than 80 % can serve as feedstock as well as wastewater containing organic

material. Above its critical point (pressure of 22.12 MPa and temperature of 374.12°C), water can reach supercritical conditions. Supercritical water gasification of biomass is mostly realized at temperatures between 600ºC and 650ºC and at a pressure of about 30 MPa. Above 600ºC, water behaves as a strong oxidant. For temperatures below 450ºC, CH_4 would be the main component in the produced gas, whereas when temperatures are above 600ºC hydrogen would be in a higher quantity [43]. The use of catalysts

can significantly improve supercritical water gasification efficiency.

- Chemical Looping gasification (CLG): The concept of CLG comes from the chemical looping combustion (CLC). The main difference is that with CLG, the objective is to produce undiluted syngas instead of heat. Similarly to CLC, the CLG process uses two separate reactors, an air reactor (AR) and a fuel reactor (FR). In the FR, biomass undergoes pyrolysis at high temperature to produce gas, tar and char, then the pyrolysis products react with the oxygen carriers (OCs) [44]. As a result, the metal oxide, which is mostly used as oxygen carrier materials (OCM), is reduced to a state of lower oxidation. Simultaneously, the OCs can serve as catalysts and accelerate the decomposition and reformation of tar. In the literature, CLG is not only associated with the utilization of OCMs for the supply of pure oxygen; but there is also a CaO-based CLG used to produce a hydrogen-rich gas by utilizing CaO for in-situ CO_2 capture [44].
- Plasma gasification: In this gasification type, electric discharges are used to ionize gas molecules in a material known as plasma. Because of the high level of energy density inside the gas along with the presence of ions and electrons, the plasma would be highly reactive. Plasma would generally be generated by using either direct current, alternating current or microwave discharge. The operating temperature inside the plasma is often greater than 5000K. Such a high temperature would facilitate the decomposition of different materials into their elementary constituents. These characteristics have given plasma gasification a special attention in the field of toxic organic wastes or some municipal solid wastes treatment for energy production.

6.3 Implications for Coal and Biomass Co-gasification Process

While coal is essentially made up of carbon, biomass on the other hand, is composed of complex compounds like hemicelluloses, extractives, cellulose, lignin and minerals. Co-gasification is thus an interesting technology because of the possibility for coal and biomass' synergism at different levels. It has been shown that it is highly beneficial to take existing gasification plants that use coal as fuel, IGCC plants in particular, and supplement them with biomass feedstock [45]. In areas where lots of biomass waste is produced, this becomes a good economic endeavor, considering supply facility and affordable costs. The mixing of biomass and coal results in a faster reaction time than coal or biomass alone, and requires fewer gasification agents (i.e. oxygen or air) to complete the reactions.

During the co-gasification process, both feedstocks will be producing syngas once they have been put into contact with the gasifying agent at a certain temperature. The process of syngas production will happen at the same time as the feedstock will go into a transformation stage but the speed of transformation (illustrated by chemical equations) will vary in accordance with the thermal, physical and chemical properties of the feedstock.

Moreover, some liquids from coal and biomass can be obtained via the co-gasification process combined with the Fisher-Tropsch technology. Unlike petroleum-derived diesel, Coal-Biomass to liquids (CBTL) can help to attenuate GHG emissions. Some research reported that the use of 30% switchgrass (Biomass) with coal for producing diesel (CBTL) using carbon capture and storage technology (CCS) produced 63% less GHG emission compared to a fossil-derived diesel [46]. There is a possibility to further decrease GHG emissions by 75% if using the more efficient capturing technique of auto-thermal reformer in CCS.

7. CONCLUSION

Gasification is not a single step process and involves many different steps such as drying, devolatization, oxidation of char and reduction in absence of oxygen. Syngas is often produced at the end of the reduction process in a gasifier. All these steps generally take place in different zones of the gasifier. For selecting a specific gasification process, the following factors are evaluated: characteristics of the coal or biomass used (such as particle size, ash content), the reactivity of the fuel, type and composition of the oxidant (air/oxygen), plant size.

In this paper, after discussing the devolatization process and char gasification reactions for coal and biomass, a comparative analysis has been made focusing on the conversion yields and the

environmental impacts. We concluded that both fuels had almost similar carbon conversion levels of about 98% at 1200ºC. It is also mentioned that conversion will increase with temperature and CO_2 concentration in the feed gas. The product gases are rich in CO as the gasification predominantly occurs through the Boudouard reaction. The CO/H_2 ratios (on N2 free basis) are approximately 38:1 and 21:1 for biomass and coal respectively at 1200ºC and 20% CO_2, using N_2 as the gasifying agent. The emission characteristics of polluting gases (detected in ppm levels) varied for the fuels. For coal, a large amount of H_2S was released whereas for the biomass, no H_2S was detected which was resulting from the nature of sulfur contained in the two fuels. Afterward, we also presented some relevant technologies which in a near future, will provide new gasification techniques with high efficiency.

These results are meaningful in the sense that they provide straightforward, insightful and relevant differences between coal and biomass gasification processes. They can serve as reference for choosing the most suitable and efficient gasification process using any of those feedstocks, but also help to better understand the behavior of gas and biomass co-gasification which is intrinsically linked to the specific process of gas and biomass gasification.

Even though gasification presents many advantages in terms of energy production, the process still faces some technical and economic challenges, mainly due to the highly heterogeneous nature of unconventional feedstocks and the relatively limited number of gasification plants worldwide which have actually achieved the expected level of technical and economic efficiency. Thus there is a need to conduct further research.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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