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POSSIBILITIES OF CO AND H₂ CONTENTS CHANGE IN THE SYNGAS PRODUCED BY
BIOMASS GASIFICATION

MOŽNOSTI ZMĚNY OBSAHU CO A H₂ PŘI PRODUKCI PLYNU Z PLYŇOVÁNÍM BIOMASY

Abstract

Thermal gasification is an advanced technology to convert purposefully grown or waste ligno-cellulosic biomass into calorific gas. Increasing the content of CO and H₂ in the product gas is important for the further use of the gas in technologies for power generation. This article describes the process of gasification and specifies parameters that determine the content of CO and H₂ in the produced gas.

Abstrakt

Termické zplyňování představuje progresivní technologii pro přeměnu cíleně pěstované či odpadní ligno-celulosové biomasy na výhřevný plyn. Zvýšení obsahu CO a H₂ v ve vyrobeném plynu je důležité pro další využití plynu v technologiích pro výrobu elektrické energie. Článek se zabývá popisem procesu zplyňování a určením parametrů, které určují obsah CO a H₂ ve vyrobeném plynu.

Keywords

Biomass gasification, biomass gasification generators, gasification reactions

1 INTRODUCTION

Concern about the global warming and climate change have led to efforts to reduce CO₂ emissions and greenhouse gas emissions by increasing the use of renewable energy and the focus is on increasing energy efficiency. Besides solar and wind energy, biomass is considered a major source of renewable energy.

The mix of renewable energy from solar, wind and biomass energy can be produced from biomass used as controllable, manageable power that will be supplied in increased quantities at a time when energy supply from wind and solar power is low.

Thermal gasification is progressive technology to convert purposefully grown or waste ligno-cellulosic biomass into usable calorific gas for cogeneration of electricity and heat with a future perspective in the production of transport fuels or other desirable commodities.

It leads to the oxidation of hydrocarbons and water vapor in the fuel and their direct reduction to flammable gases, distillation products and mineral residue at the gasification of biomass thermal chemical processes. The process is controlled by a controlled supply of the gasification agent and the necessary heat of reaction.

The product of the gasification processes a gas containing calorific components (H₂, CO, CH₄ and other minor compounds), accompanying components (CO₂, H₂O, N₂) and impurities (tar, dust, sulfur compounds, chlorine, alkali and others) [1]. Just increase the calorific content of gas

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components such as CO and H₂ in the product gas is important for the further use of gas in technologies for power generation.

2 BIOMASS

Biomass is the fourth most widely used energy source in the world for oil, coal and natural gas. Energy utilization of organic matter (biomass) is limited by their low energy density, high local emissions of polluting substances [2], and complexity of the supply chain, which must compete with the main use of organic substances thus food (or material use).

Using organic waste as feedstock for high-efficiency cogeneration gasification plants would solve all the above mentioned disadvantages associated with the use of biomass as an energy source. For selecting the forms of biomass feedstocks which is to be used in the gasification process, as the first criterion is needed to consider the availability of this form of biomass in a larger scale (t / year) in the area.

Biomass density significantly affects any handling thereof and its storage, when the need for a large storage space that must be covered in order not to increase the moisture stored biomass.

In the purpose-grown biomass such as fast-growing poplars and willows, is due to obtain fuel with the lowest possible moisture content performs "harvest" outside the growing season when plants have the lowest moisture content. For example, in plantations of fast growing trees are harvested in frost.

Energy conversion in gasification consumes energy. Therefore, the input fuel preferred fuel with high calorific value and energy density. This means that the biomass with lower humidity is preferable, because the moisture decreases the calorific value of biomass. Reducing the moisture content can be achieved by free drying in air or by using the surplus heat produced during power generation. In this case, you can take advantage of biomass with 50% humidity.

While black and brown coal is typically only 20% volatile matter and the remaining 80% is non-reactive coke (fixed carbon), biomass accounts for more than 80% volatile matter, which goes well gassing, the remaining 20% is charcoal (fixed carbon).

Chemical composition (C, H, O, N, S, Cl) is another important aspect which should be considered [3], [4]. For lignocellulosic biomass, the chemical composition (on a dry ash-free sample) is generally more stable than other solid fuels (municipal solid waste, coal).

Biomass has generally very low content of sulfur and chlorine in comparison with coal and municipal solid waste. To utilize the produced gas (eg. Fuel cells) it is mostly necessary to clean gas from of these compounds to the level required by the specifications of the technology.

Finally, the content of ash and tar is one of the main obstacles economically viable applications of biomass gasification. Fuel with a high ash content requires more attention, because the ash leads to problems with its sintering, sticking to the generator area, erosion and corrosion problems. The fact that the ash carried away with the produced gas, the gas complicates cleaning procedures.

Tar condensation at high temperature causes clogging and damage to the downstream devices. When we summarize the above facts, the most appropriate biomass for gasification must be available in large quantity (t / year), have good physical properties (low water content and high bulk density) and chemical properties (high caloric value, high volatile content, with low ash content, high ratio of carbon to nitrogen content, low content of chlorine and sulfur).

As mentioned above, attention should be paid to the biomass waste, such as:

- purpose-grown energy crops (eg. Miscanthus),
- agro-industrial residues (eg. Exhausted olive),
- shells (eg. hazelnuts, walnuts and almonds),

- pruning (eg. Beech, oak, spruce, poplar, willow),
- straw (e.g. wheat, corn, rye, barley).

It is important for gasification in a fluidized bed, so that there was a good mixing between the fuel (biomass) and the fluidized bed material. Therefore, the biomass density should be comparable with the density of the bed material. Another important parameter which must be considered is the size and shape of the biomass in terms of traffic to the gasifier. To ensure stable and efficient gasification of biomass must be processed to a uniform size and shape, and be supplied to the gasifier stepless speed. This can lead to significant costs for crushing (chip size 1-2 cm is currently a good compromise).

Table 1 shows the physic-chemical properties of these different types of biomass. The table shows that the shells have preferable characteristics (low moisture content that does not change much, high density, low ash content, high calorific value). Prunings has more fluctuating characteristics. Straw has not only very volatile nature, but also a higher ash content, which in many cases has a lower melting point than the temperature of the gasification, which causes clogging of the generator. As for the content of carbon, hydrogen and oxygen, lignocellulosic biomass has almost the same percentage (respectively 41-51% carbon, 5-6% hydrogen, 36 to 44% oxygen). N, Cl, S is very low percentages that vary depending on the type of biomass and on the growing conditions (soil, fertilizers, etc.).

Tab. 1 Physic-chemical characteristics of biomass [5]

Biomass type	LHV (MJ/kg dry)	Fixed carbon (% wt)	Volatile matter (% wt)	Ash (% wt)	C (%)	H (%)	N (%)	O (%)	Cl (%)	S (%)	Humidity (% wt)	Density (kg/m ³)
Miscanthus	18	19	71	5	45	5	0,5	40	0,2	0,08	4	240
Exhausted olive	20	19	77	4	51	6	0,3	38	0,02	0,02	9	350
Shells	18-20	20-25	74-78	1-2	48-51	6	0,2-0,5	41-44	0,02-0,03	0,01-0,03	11-14	300-500
Pruning	16-18	12-20	70-85	0,5-4	45-49	5-6	0,1-0,8	36-44	0,01-0,08	0,01-0,08	7-25	200-300
Straw	15-18	16-18	67-76	5-15	41-47	5-6	0,3-6	36-44	0,03-0,4	0,04-0,2	7-12	20-140

3 DESCRIPTION OF GASIFICATION PROCESS

Gasification is a process of chemical transformation of carbonaceous fuels into combustible gases through the free or bound oxygen at high temperatures. The free oxygen is supplied to the process in the form of air, oxygen enriched air or pure oxygen. Chemically bound oxygen is usually fed in the form of water vapor or carbon dioxide [6]. Gasification process can proceed in two ways:

- with partial combustion of fuel supplied to the process (autothermal process),
- with external supply of thermal energy to the generator (allotherm process).

Biomass gasification is a complex process in which it runs a lot of thermochemical reactions. Their goal is to produce energetic gas. Biomass gasification takes place at substoichiometric conditions ($0.2 < \lambda < 0.4$) in the overall balance of the process. The pyrolysis process and the combustion take place locally at the same time. A very important stage - dry (drying zone) occurs in terms of the energy balance at the beginning of the process of biomass gasification. It is important from the standpoint of balance of energy which must be supplied to the process to the evaporation of moisture from the fuel. Higher moisture content in the fuel consumes more power you need to create in the process, which has a negative effect on the calorific value of the gas generator - it will be less calorific. Water vapor arising from the drying of fuel contaminates the gas of from 10 to 50% of its

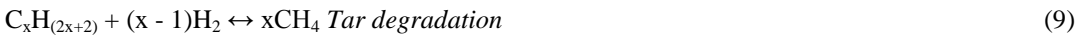
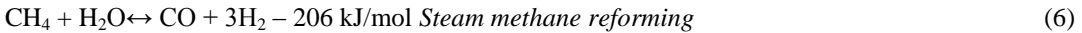
volume. One of the positive effects of the water vapor is its reaction with carbonisation product in the reduction zone, the result of this reaction is the production of H₂ and CO.

3.1 Reactions of gasification process

In the gasification process, there are numerous types of reactions which are dependent on various parameters. The final composition of the gas is the result of sequential and parallel reactions, the substrates supplied to the process. In this process we are able to distinguish between homogeneous and heterogeneous reactions. Reactions in the process can also be divided into endothermic and exothermic reactions.

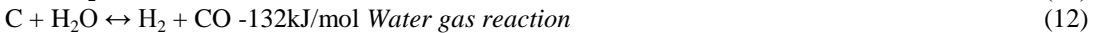
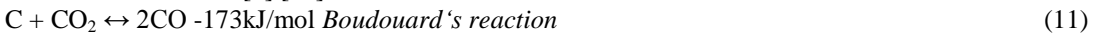
Homogeneous reactions of gasification process

Homogeneous reactions are reactions between the compounds in the same condition state. In the gasification process, homogeneous reactions occur between the gaseous products from the drying zone, pyrolysis and oxidation [7]. Homogeneous reactions are in most cases exothermic reactions. Reactivity of individual components and the energy of chemical bonds formed affects the amount of energy and the resulting product composition. Energy generated during the oxidation reaction is necessary for release of volatile matter and active tars [8]. To typical homogeneous reactions we may include the following:



Heterogeneous reaction of gasification process

Reactions which occur between the substances in different physical state, called heterogeneous. The main reactions between carbonisation products and gaseous products formed in the zones of drying, pyrolysis and oxidation. Their occurrence speak of the quality of the gasification process and also the uniform access of oxidizing agent and reactant gases (CO₂, H₂O) to the solid carbon. The following are the major heterogeneous reactions indicating the amount of energy consumed or released [9] [10].



3.2 The equilibrium constant of a chemical reaction

Equilibrium constant of chemical reactions determines the state of the chemical reaction between the substrates and products involved in chemical reactions. This state is dependent on temperature and pressure in the reaction surrounding. Equilibrium constant of chemical reactions is determined by the speed and direction of the reactions. Theoretically, the reaction can take place under suitable conditions in both directions. If the speed of the reaction for products is higher than for substrates will convert substrate to product and reactions will run to completion. It is therefore irreversible. If the reaction rate is the same in both directions, it leads to formation of chemical equilibrium. At equilibrium state, the number of substrates and products remain unchanged, although both reactions also take place. Below is a statement of chemical equilibrium constants of Guldberg-Waag:



From the above formula it can be deduced that there are two rates of reactions: k_f forward, backward k_b . Then the rate of loss of components (e.g. A), the reaction will be expressed as follows:

$$d[A]/dt = -k_f[A]^n[B]^m + k_b[C]^p[D]^q \quad (22)$$

What you see is the resulting rate of one and the other side. If the reaction rate is the same in both directions, from a microscopic point of view at any given moment a certain amount of product and the same amount of product reacts on the substrate. From the macroscopic point of view, however, a whole seems calm. The presence of reactions is evident at the molecular scale.

Equilibrium state can be written as:

$$(dA/dt)_f + (dA/dt)_b = 0 \quad (23)$$

Condition for reversible chemical reaction to equilibrium:

$$[C]^p[D]^q/[A]^n[B]^m = k_b/k_f = K_x \quad (24)$$

Theoretically all chemical reactions have reached equilibrium when they take place over a sufficiently long period. This means that for each reaction, there are conditions under which the reaction is reversible [11]. In practice, in many cases it does not create an equilibrium for various reasons:

- The product (or one of the products), of the reactions may be removed from the system faster than the reaction reaches equilibrium. Reaction continues until the end in one direction only.
- Substrates and products are present in two immiscible phases, either reactions takes place at the interface of the individual substances very slowly (the reaction rate is limited by the amount of substances), or there is no response at all due to the effective separation of the reactants.
- In the case where the reaction has a small time scale of observation.
- One of the reactants is continuously added to the system from outside, and then the system cannot achieve equilibrium.

The concentration of reactants in the equilibrium does not depend on the mechanism of the reaction, but only on thermodynamic conditions such as temperature and pressure. Catalyst does not affect the amount of reactants in balance, but has a big impact on the time at which this state will be achieved.

Influence of temperature and pressure on the equilibrium constant of chemical reaction

Equilibrium constant K_x depends on the temperature and pressure of the process. These relationships were described by Van't Hoff:

$$\left(\frac{\partial \ln K_x}{\partial p}\right)_T = -\frac{1}{RT} \sum \nu_j \left(\frac{\partial g_j}{\partial p}\right)_T = -\frac{1}{RT} \sum \nu_j v_j = -\frac{\Delta_R v}{RT} \quad (25)$$

$$\begin{aligned} \left(\frac{\partial \ln K_x}{\partial T}\right)_p &= \frac{\partial}{\partial T} \left(-\frac{1}{RT} \sum \nu_j g_j\right) = \frac{1}{RT^2} \sum \nu_j \left(g_j - T \frac{\partial g_j}{\partial T}\right) = \frac{1}{RT^2} \sum \nu_j (g_j - T s_j) = \\ &= \frac{1}{RT^2} \sum \nu_j = \frac{1}{RT^2} \sum \nu_j i_j = \frac{\Delta_R i}{RT^2} = \frac{q_p}{RT^2} \end{aligned} \quad (26)$$

Van't Hoff's equations allow you predict what changes of pressure or temperature affect achievement of equilibrium reaction. If K_x grows, the value of molar components x_j increases, which correspond to the stoichiometric coefficients. Increasing the pressure advances the state of chemical equilibrium in the direction in which the individual components of the reaction mixture achieves a smaller specific volume or a higher density. [12] This is very important for gas phase reactions.

3.3 Kinetics of gasification process

The kinetics of the process of simultaneous gasification of coal and biomass is the subject of many research works. Their aim is to describe the characteristic phenomena occurring in the gasification process in dependence on time. The complexity and variety of homogeneous and heterogeneous reactions has an effect that the creation of a theoretical simulation model that is closer to real conversion process of fuel, it is very difficult [13], [14].

For a theoretical definition of the kinetics of reactions in the conversion process is necessary to know reaction described by equation Waag-Guldberg's (21) and reaction speed (22) set out in section 3.2. As previously defined, the reaction rate is the change of concentrations of substrates and products in time. Equation (21) describes the reaction substances $A + B$, in which $C + D$ rise. During the reaction there is a loss of one of the reactants (27).

$$w_r = \frac{C_{A0} - C_{A1}}{\tau_0 - \tau_1} = -\frac{dC_A}{dt} \quad (27)$$

Comparison of the kinetics of different fuels such as coal, biomass or oil, gives very different results. In the gasification of coal or coke that have little or zero volatile matter, it is very difficult to ensure the contact of oxygen with carbon. Gasification of oil which is composed of the volatile matter almost 100%, it will take place in the gaseous phase in a different way. The proportion of volatile matter in the biomass is also very high, but it depends on the ash content. The proportion of ash in fuels from biomass can be up to 20% of mass. Variability of this parameter influences the change of process conditions and participation of homogeneous and heterogeneous reactions in the process of gasification. Low melting point, which depends on the composition of the ash, also determines the nature of reactions. In describing this relationship for heterogeneous Boudouard's equation (11) is determined the degree of carbon conversion:

$$r_m = \frac{dC}{dt} = k_m \cdot C_{CO_2} \quad (28)$$

It consists of the concentration of CO_2 in the gas and the value of the equilibrium constant k_m . According to the Arrhenius law, establishing the influence of the temperature at which the reaction takes place, it is possible to express this relationship also has the following formula:

$$k_m = A \cdot e^{-\frac{E_A}{RT}} \quad (29)$$

Reaction rate constant and the activation energy E_A are called kinetic constants. The constant A is also known by symbol k_0 and called constant frequency. The unit of constant is s^{-1} , but it can vary depending on the unit used and the concentration of timescale.

4 TYPES OF GASIFIER TECHNOLOGY

Gasifiers can be divided into two main groups [15]:

- with fixed bed (from which is derived a moving bed gasifier),

- with fluidized bed.

Within the fixed bed gasifiers can be distinguished updraft when biomass progresses downward and made gas passes from the bottom upwards; downdraft, when the produced gas and the biomass move in the same direction from the upper to the lower portion of the generator; with crosscurrent, when the biomass moves downward, and the produced gas is discharged at a right angle.

4.1 Fixed bed gasifiers

Biomass moving downward is dried and pyrolysis in the updraft of the generator, whereby the resulting char continues to move downwards, where it is gasified. The pyrolysis vapors are guided upward by the upward flow of hot gas produced. Vaporized tars in the gas either condense on the cold descending layer of fuel, or are removed from the space of the generator together with produced gas. Due to the extremely high content of tar in the product gas, the construction of the generator is suitable for use in combination with the direct combustion of the product gas in the burners.

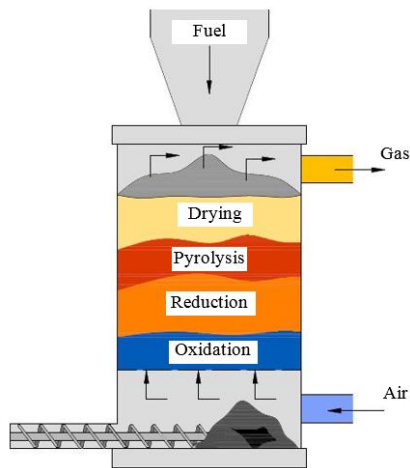


Fig. 1 The updraft generator [1]

Biomass is, together with the gasifying medium forced to pass through the constriction (throat) in a downdraft gasifier. In this constriction occurs most gasification reactions. The high residence time of the biomass results in a high conversion of charcoal gas. Given that the gases leave the gasification unit at temperatures of about 900-1000 ° C [1]. The overall energy efficiency of a downdraft gasifier is low (due to high content of heat dissipated in the hot gas). Downdraft gasifier is generally used for the production of electric energy in combination with the internal combustion engine. Downdraft gasifiers are not suitable for use in the higher outputs (> 1 MW), because they do not allow uniform distribution of flow and temperature in the narrowed area (throat).

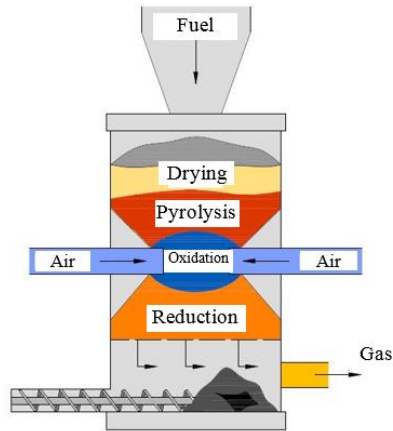


Fig. 2 The downdraft generator [1]

Crosscurrent gasifier is downdraft generator with a moving bed, in which fuel is fed from the top and the air is injected from the side through a nozzle. One of its important features is the relatively small reaction zone with low thermal capacity, which provides a faster response time than it is possible for any other type of generator a fixed bed.

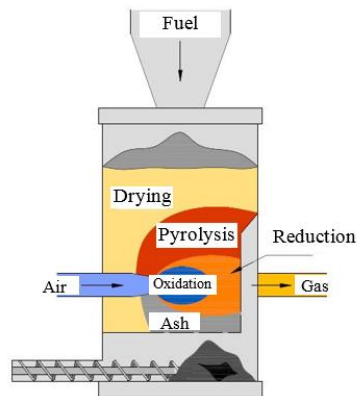


Fig. 3 The crosscurrent generator [1]

4.2 Fluidized bed gasifiers

In the gasification in a fluidized bed the solid fuel is mixed with the hot fluidized bed material (inert sand, ash, catalyst) and it is maintained in fluidized stream of gasifying medium [16].

Due to the intense mixing of gasification agent with the fuel stream, the process is not divided into areas having prevailing reactions of one type - as is the case with generators fixed bed. All the processes take place simultaneously at each location of the fluidized bed. Only the place of supply of the gasification agent (just above the grate), we can identify an area where oxidation reactions dominate over reduction reactions. For these reason is conversion of biomass in a fluidized generator almost 100%.

For the same reasons, fluidized gasifiers are generally more tolerant to the input material and suitable for large installations. This a change compared with fixed bed gasifiers, which are quite sensitive to the quality of raw material (uniform material, accurate humidity etc.).

Fluid generators work with uniformly distributed and relatively low temperatures (900 ° C) [1], which means that they do not need moving mechanical components which could resist the high temperatures, because the perfect mixing is guaranteed by fluidization. They can without any problems gasify fuels with a low ash melting point, it means also fuels with high ash content can be gasified.

Among the disadvantages of fluidized bed gasifiers include difficult controllability of performance and the abrasive effect on the walls of gasifier due to the rapid movement of the fluidized bed material.

Selected characteristics of the fluidized bed in dependence on the gas velocity in the free cross section of the generator are in fig. No. 4. Speed of gas and solid particles is proportional to the particle size for a given power generator. Most devices has a velocity of the gasifying medium 5 to 30 times greater than the fluidization velocity threshold [1]. For each particle size it is necessary to select an optimum hydrodynamic regime.

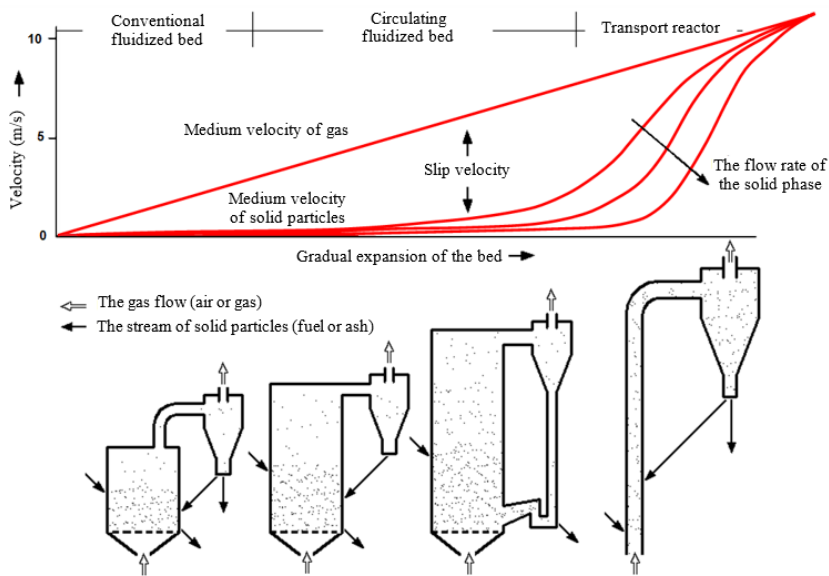


Fig. 4 Types of fluidized bed gasification [17]

Tab. 2 Parameters of gasifiers with fixed and fluidized bed [18]

Type of generator	Fixed bed	Fluidized bed
Feedstock size [mm]	10-100	0-20
Maximum ash content [wt. %]	< 6	< 25
The residence time of gas in the generator [s]	1	1
The residence time of char in the gasifier	< 1 hour	0 – 10 min
Operating temperature [°C]	800 - 1400	750 - 950
The ratio of max. and min. output of device	4:1	3:1
Thermal output [MW]	< 2,5	1 – 50
Start-up time	Minutes	Hours

6 CONCLUSION

The main parameter that affects the content of CO and H₂ in the synthesis gas that is the gasification (oxidizing) medium, which may be air, pure O₂, steam, CO₂ or a mixture thereof as shown in Tab. 3.

Tab. 3 Synthesis gas composition for each oxidant [19], [20]

Oxidant	N ₂	CO ₂	CH ₄	CO	H ₂
	[%]	[%]	[%]	[%]	[%]
Air	56-59	14-17	2-4	12-15	9-10
Steam/CO ₂	-	10-19	5-12	30-45	24-50
Oxygen	-	25-29	4-6	30-37	30-34

Due to the easy availability and null costs, air is most commonly used as a gasification agent. However, it contains a large amount of nitrogen that lowers the calorific value of the produced syngas. Furthermore, it requires a larger volume due to the higher output auxiliary devices gasification technology.

Steam as a gasifying medium increases calorific value and the H₂ content in the synthesis gas and can be made with almost no cost for water, using an excess of produced heat energy [21]. Calorific value and the content of H₂ and CO in the synthesis gas increases the use of CO₂. Increasing the H₂ content (over 60%) and CO₂ (10 to 30%) reduction of the CO content (35-10%) and a slight decrease of CH₄ when the ratio S / B (steam a biomass ratio) was increased to 0, 5 – 2,5, it was shown by Herguido et al. [22] in his work. But using CO₂ increases operational costs because of the relatively expensive and complex manufacturing of pure CO₂. Use of steam or CO₂ as a gasification agent supplies heat required for the endothermic gasification reactions. This can be done allothermally, by circulation of the hot material, or using heat exchangers. In this case, the combustion products do not come into contact with the product gas which thus has a higher calorific value. The second way is autothermal heat supply, air supply, [23] or O₂ [24] to the gasifier for heat generation by partial combustion of biomass.

Using pure O₂ we avoid increasing the nitrogen content in the synthesis gas, which increases its calorific value. Use of O₂ as a gasifying medium, like CO₂, reduces its high production costs.

Other factors that affect the content of CO and H₂ in the syngas, include the physic-chemical composition of the fuel (ie biomass feedstocks, see Tab. 1), the design of the generator and the residence time of fuel in the reduction zone. Converting exhaust gas from the oxidation zone (CO₂ and H₂O) to H₂ and CO occurs in the reduction zone.

The CO and H₂ content in the syngas can be increased by also using nickel catalysts [25], which are among the so-called synthetic catalysts. Production of nickel catalysts, however, requires high cost and is sensitive to deactivation by sulfur compounds. Additionally catalysts reduce gasification temperature, which leads to lower heat losses. The advantage of this group of catalysts is almost total reduction of the concentration of tars from the gas at a temperature of 900 ° C. [25]

In order to obtain higher content of CO and CH₄ in the synthesis gas, CO₂ is separated in larger gasification installations which is fed to the reduction zone. [26]

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