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Methods to improve the performance of fluidized bed biomass gasifiers

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Abstract

In fluidized-bed gasification (FBG) of biomass and waste the temperature is maintained low enough to avoid agglomeration. This decreases carbon conversion and generates a tarry gas, reducing process efficiency and the gas utilization. In the present work optimization of FBG to achieve high char and tar conversion in the reactor is studied. Staged gasification is identified as a rational way to achieve the target with reasonable complexity and cost, so it is an ideal method for power production in small/medium biomass gasification plants. A recent development based on a tree-stage fluidized bed gasification system, is presented.

Keywords

Gasification, fluidized bed, char, tar, biomass, optimization

Introduction

Gasification is a thermochemical route for conversion of solid fuel into a gas, which can be used in a variety of applications [1,2]. Gasification in a fluidized bed (FB) has several advantages over that in fixed/moving bed and entrained-flow gasifiers [3]. The FB provides high mixing and reaction rates, accommodates variation in fuel quality and allows scaling-up of the process. Various concepts have been developed for gasification in FB. Stand-alone, air–blown, bubbling FB gasification (FBG) is the simplest, directly heated design, but it delivers a gas diluted by nitrogen, having low heating value (4-6 MJ/Nm^3) and high tar content (10–40 g/Nm^3). Medium heating-value gas (12–15 MJ/Nm^3) can be produced by directly heated gasifiers using mixtures of oxygen and steam in one single reactor [4], or by indirectly heated gasifiers using air and steam in a twin bed reactor [5]. In all types of FBG the tar concentration in the gas is high, since the process temperature is kept relatively low to prevent agglomeration and sintering of bed material [1–3]. This results, in addition, in incomplete carbon conversion in directly heated FBG.

The unconverted carbon in the char reduces the efficiency of the process and the high concentration of heavy tars limits the application of the gas to direct thermal applications, like burning in kilns and boilers. Therefore, applications, such as gas engines, turbines, and fuel cells, and conversion of the gas for synthesis of fuels or chemicals, need extensive and costly gas cleaning [1]. Effective methods to capture tar downstream of the gasifier are available [6–11] based on physical separation and condensation (wet/physical methods) and on reforming/cracking of the tars in the hot gas. Wet methods have been tested, using water [6,7] or organic solvents [8], and have been reported technically efficient. This way to clean the gas,

however, is too complex and expensive for small or medium-size plants [1]. The conversion of tar by catalytic reforming/cracking in a downstream vessel is also efficient [9–11] but catalysts have technical shortcomings. Novel catalysts, tested in the lab, come up daily but they need demonstration, so there is no commercially available catalyst yet [12]. In summary, reliable and relatively cheap methods to reach high char and tar conversion are needed, especially for small to medium scale plants. An interesting option to achieve such target is to convert the char and tar in the reactor [13] (primary methods), avoiding difficult secondary cleaning methods.

Fundamentals on fuel conversion process in FBG

A biomass particle fed to the reactor undergoes a series of conversion processes. Initially, drying, devolatilization and primary pyrolysis yield char and volatiles. Subsequently, the volatiles and char may be oxidized, and finally, the char may be gasified by carbon dioxide and steam. Fuel particles shrink, and primary fragmentation may occur immediately after the injection of the fuel into the bed. Secondary fragmentation and attrition of char take place together with char conversion. The volatiles include non-condensable gases, such as CO_2, H_2,…, condensable gases (tar), and water (chemically bound and free water).
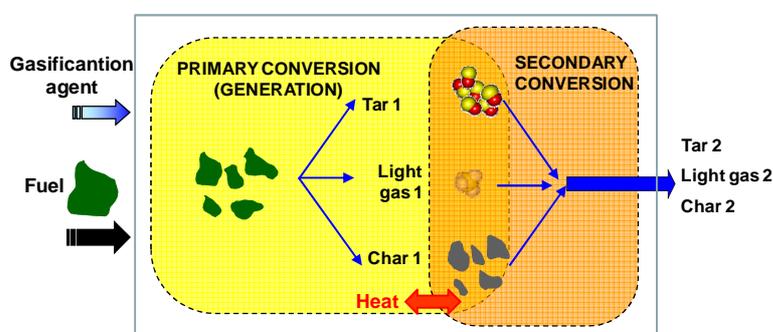

Figure 1: Scheme of reactions of the primary conversion process during devolatilization.

The distinction made in Figure 1 between primary and secondary conversion is based on the different times of conversion of the various processes [3]. The rates of char gasification with H_2O and CO_2 are orders of magnitude lower than those of primary pyrolysis: it takes a few seconds to devolatilize 90% of a fuel particle whereas a few minutes are required to gasify 80% of char with steam at temperatures below 900°C. On the other hand, conversion of volatiles is rapid if they react with O_2, but much slower if they react with steam. Due to the fast release of volatiles, the gasification agent does not penetrate into the particle during devolatilization. Therefore, the rate and yield of devolatilization is quite insensitive to the composition of the surrounding gas, and the bed simply provides the heat for the process [3]. Once the volatiles are emitted from the particle, they mix with the surrounding gas and react by secondary reactions, mainly reforming and oxidation, resulting in secondary gas, char and tar, indicated with "2".

The conversion of char and tar is related to the effective time of reaction with the gas and catalyst, which in turn depends on the residence time of the fuel and char particle in the bed and on the local conditions of mixing in the reactor [14,15]. Of particular importance are the contact between char and tar with oxygen and steam and the position where the fuel particle is devolatilized in the reactor. Formation of bubbles, bypassing of gas, entrainment of material and other factors influence the reaction time [3,15].
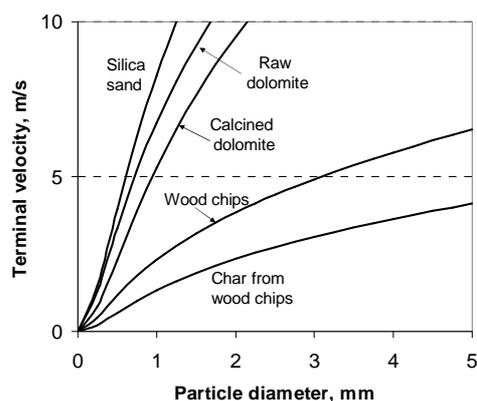
Figure 2: Terminal velocities of single particles of various materials in a CFB (calculated for air, at 850 $^{\circ}$C and at a pressure of 1.1 bar) as a function of particle size. After [3]

The key operation parameter in an FB is the superficial velocity of the gas and the properties of the fuel particle (mainly size and density), determining the degree of mixing and entrainment in the reactor [3,16]. High superficial velocity improves solids mixing, but biomass particles with lower density and larger size than bed particles can still be non-uniformly distributed. At a given fluidization velocity, char particles more likely circulate in the bed, while devolatilizing fuel particles tend to float on the bed´s surface because of the lift force of escaping volatile matter [3]. On the other hand, increasing gas velocity enhances entrainment of material and decreases the residence time of a particle in the reactor. A qualitative judgment of the tendency of a particle to be carried away can be made by the terminal velocity of a single particle u_t. Figure 2 presents u_t versus particle diameter for different materials used in a circulating FBG [3]. At a superficial gas velocity of 5 m/s, the diagram indicates that, while silica sand of 0.5 mm would stay in the bottom bed, dolomite of 0.6 to 1.0 mm would remain in the bed initially, but once calcined, it could be rapidly elutriated. Biomass like wood chips with particle sizes smaller than 3 mm would be carried away from the bottom bed, whereas the char generated from this fuel with particle sizes up to 10 mm could be transported. Recirculation yields higher carbon conversion, because it increases the residence time of particles. However, there are circumstances when the effect of recirculation is reduced because the reactivity of the char falls due to deactivation by interactions with volatile compounds [25]. In circulating FBG, large devolatilizing fuel particles move towards the top of the riser, increasing the tar yield of the gas. In a bubbling bed, in contrast, the fuel particles are likely to remain in the bed (or on its surface) most of the devolatilization time due to lower superficial velocity (lower entrainment). Entrainment of biomass particles during devolatilization does not play an important role in a bubbling unit but entrainment of finer char particles may be severe.

The rate of devolatilization and the yields of primary char, tar, and light gases vary mainly with temperature and heating rate at which the fuel is transformed. These factors are in turn determined by the fuel particle size and the operating temperature of the reactor [17]. The rate of devolatilization of mm-size particles in an FBG is usually controlled by internal heating [3,17]. The fuel emits a variety of organic compounds, from aliphatic chains to parent fuel structures. Vapour-phase secondary pyrolysis in the internal pores of the fuel particle involves complex parallel and consecutive reactions, included within the primary pyrolysis [17]. The compounds released are thermally unstable at temperatures over 600°C due to the multiple aliphatic bonds and heteroatoms in its structure. Most tars are emitted before the particle is heated above 600°C [3], but outside of the fuel particle they meet the bed temperature and their chemical nature changes by breaking of the aliphatic structures into lower hydrocarbon molecules and light gas, as well as by rearrangement of lower hydrocarbon chains to more stable aromatic structures. In general, the secondary reactions decrease the tar yield but enhance generation of refractory non-substituted, stable, polyaromatic hydrocarbons (PAHs)

[18–21]. At temperatures below 900°C, PAHs are not significantly converted into gas via non-catalytic steam reforming in the time range available (seconds).

Conversion of char generated after devolatilization (secondary conversion) is governed by the chemical reactivity, secondary fragmentation, and elutriation of char fines. In a directly heated FBG the char is not much affected by oxygen, since the oxygen is preferentially consumed by the volatiles, and the contribution of combustion to the overall char conversion is small. Then the char is mainly converted by CO_2 and H_2O, which are slow reactions [22]. The rate of reaction of char with different gases can be represented by global nth order kinetics, valid under certain operation conditions [3,22]. More complex Langmuir-Hinshelwood kinetics are necessary for analysing a broader range of conditions [22]. During char conversion by gasification in FBG, the reaction zone extends over most of the particle, and the interior surface changes significantly. The overall conversion in the bed depends also on the distribution of char particles in the bed, which, besides the char properties, is mainly governed by solids mixing, and so, by the gas velocity [3].

General trends of devolatilization can be obtained by comparison of published data, as it is shown in Figure 3, where the yields of char, light gas, and liquids (tar + water) as a function of the temperature at various heating rates (particle size plus heating rate of the equipment) are presented for a variety of biomass materials [23]. At the lowest temperatures (< 300°C) char is the main product. At middle-range temperatures (450-550°C) a maximum is observed for liquids, being secondary the influence of heating rate. On heating up to around 450-550°C, slow heating rates give more char and less tar than fast heating rates due to intraparticle charring of the primary tars and the low activity of secondary reactions of volatiles. The observed decrease in the yield of char as temperature increases indicates that the major mass loss of fuel occurs in the range of 200-600°C. At these low temperatures, the heating rate has a small influence. As temperature increases above 450-550°C the variation in the yield of char is small, with low heating rates generally associated with higher yield of char. The influence of the heating rate on the tar yield becomes less important at the highest temperatures analyzed (> 800°C). The low temperature range of the thermal decomposition of biomass can be referred to as the primary pyrolysis stage, which is the likely process to apply to mm-sized particles in FBG. It is worth to say that primary fragmentation has not usually taken place in the tests included in Figure 4, but it can occur in real FBG [3,24], changing the effective particle size to be devolatilized (primary fragmentation can be considered almost instantaneous).
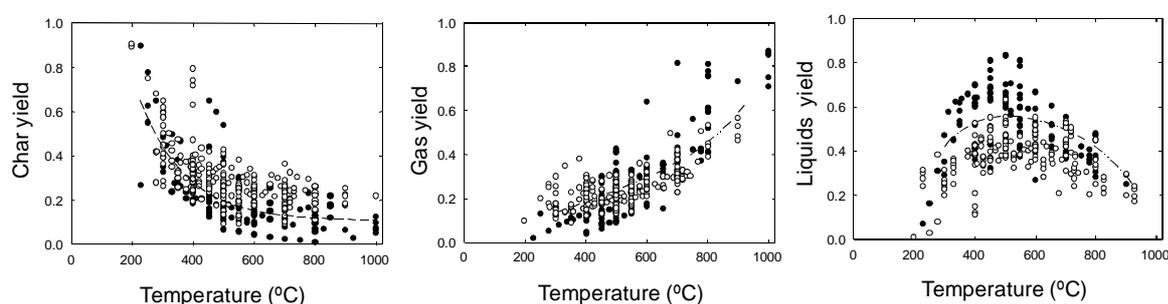

Figure 3: Yields of char, light gas and total pyrolytic liquids (tar +water) as a function of the peak pyrolysis temperature. ● -"fast heating rate"; ○ -"slow heating rate"; ··—·· empirical model from [23]

Methods to improve char and tar conversion in conventional FBG

Primary methods are discussed here only since complex secondary gas cleaning is not feasible for small to medium gasification plants for electricity production. An ideal way to gasify should eliminate the need for further complex gas treatment and the char should be completely

converted in the gasification reactor. For a given type and flow rate of biomass (thermal input), the optimization can be made by adjusting the flow rate of oxygen, steam (and catalyst or additive, if any) to give sufficiently high temperature, residence time of the char and tar (gas mixing and effective time in contact with catalyst and char), under the condition of a safe operation (without sintering) [1,3]. In the following, various ways will be illustrated to optimize the gasifier by adjusting composition and flow rate of gasification agent, staging of the gasification agent, and addition of in-bed catalyst.

Figure 4 summaries the effect of temperature in a gasifier on the key output variables: char conversion, tar concentration and heating value of gas, as well as sintering. It is seen that raising the temperature increases tar and char conversion. However, the danger for sintering of ash and bed material also increases and sets the maximum temperature. Considering the balance between benefits and drawbacks associated with the thermal level, the temperature range of a biomass gasifier is between 800 to 900 °C. Even at the highest temperature acceptable, it is difficult to convert PAH into gas via steam reforming by non-catalytic reactions or by contact with bed material. As discussed previously, mass transport resistances provoked by fluid-dynamic effects may limit the effective contact of tar and catalyst.
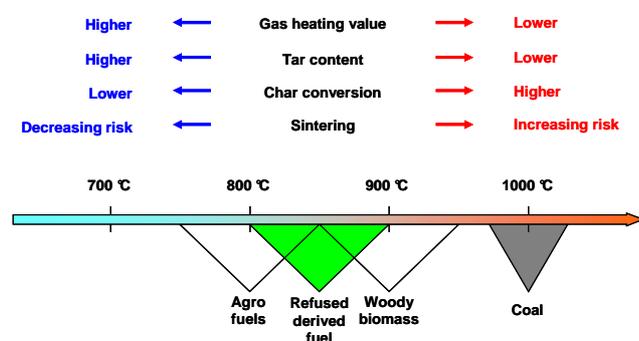

Figure 4: Effect of temperature on parameters and processes during gasification [13]

Figure 5 shows the distribution of the fuel energy into sensible and chemical energy of the gas as a function of the oxygen addition (quantified by the stoichiometric ratio, SR). The chemical energy in the gas increases with SR up to a certain level because more volatiles are emitted and more char is converted with raising temperature. This occurs from a stoichiometric ratio of 0 up to 0.3-0.4, because the fuel is increasingly converted. However, when the oxygen supply exceeds the point where the char has already been completely converted into gas, more fuel is burned to CO_2 and H_2O and the heat release increases at the cost of product gas, lowering the chemical energy in the gas. An optimum is found where the chemical energy in the gas is maximized with significant tar reduction. The optimum from the tar point of view, however, is not clear, since the reduction of tar is not the key issue: the dew point of the gas is a more correct index of gas quality [8,25]. The calculations in Figure 5 were made for equilibrium. In practise, equilibrium is not attained because the conversion of char controls the process and the optimum (complete char conversion) is shifted to higher SR.
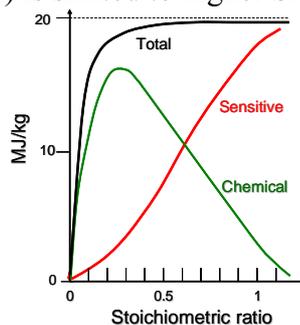

Figure 5: Energy conversion from solid fuel to gas.

Figure 6 shows the effect of residence time on the char conversion attained in an FBG at various temperatures using a model that takes into account the fluid-dynamic and fuel-conversion processes affecting char conversion [3]. The residence time of the char is the ratio of the mass of char in the gasifier to the fixed carbon of the input fuel-stream. The operational data correspond to a 1m-internal diameter atmospheric air-blown bubbling FBG [26] processing wood pellets. It is seen that at 800ºC the residence time of the char has to be longer than 0.5 hours to achieve a char conversion of 80%. In contrast, at 900ºC, the conversion is 10 times faster. The points in the graph correspond to different air-biomass ratios and thermal input of the gasifier, hence, the superficial gas velocity. A similar figure for the conversion of tar in the bed as a function of residence time is not possible because there is no reliable tar conversion model yet, considering key effects such by-passing of tars with the plumes of volatiles, interaction between one tar and another during conversion, etc.
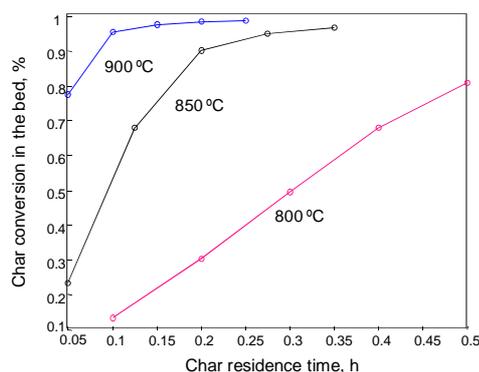

Figure 6: Effect of temperature on char residence time on char conversion in a FBG. Calculated by modeling [3]

Addition of steam to a directly heated gasifier enhances tar reforming and char gasification, improves the quality of the gas, and reduces its tar content [27,28]. However, steam addition reduces the temperature of the gasifier and more oxygen has to be added to maintain the temperature level, lowering the heating value of the fuel gas produced [28]. There is an optimal steam to oxygen ratio where steam addition positively compensates for the burnt fuel gas because char is further converted, producing CO and H_2 [27,28]. Steam addition at high temperature is an effective measure [29] but the temperature of the input steam is limited to that achievable by heat integration (for instance by heat exchange with the produced gas) if the gasification process is conducted autothermically (no external heat is added). Low-cost oxygen, such as produced by membranes with a purity of 40-50%, together with steam, preheated by the hot produced gas, has been shown to improve the process significantly, achieving a carbon conversion of up to 97% [28]. However, the tar content in the gas is not reduced to the limit required, and the dew point of the gas is above 100°C.

Staging of the gasification agent makes it possible to create various thermal levels in the gasifier. The principle is illustrated in Figure 8(a). Staging by injection of secondary air has been tested in conventional FBG at pilot scale [30–32]. A portion of the inlet oxygen (air) is conducted to a port situated in the upper part of the bed or the freeboard. Significant reduction of tar has been reported (below 1g/Nm^3) [31]. These data, however, should be interpreted with caution: in [31] the air ratio is increased simultaneously with the re-distribution of the air, i.e. more gas is burnt, lowering the heating value of the gas; in [31] a great reduction is achieved, but the operation is conducted with an external oven so it is not possible to make any conclusion about the tar reduction under autothermal operation. Injection of secondary air at constant air ratio, i.e. to analyze the effect of injection in an isolated way, has been shown to be effective for reduction of phenols and other light tar compounds [32], but the total tar concentration is still high (a few grams per Nm^3) and the proportion of stable aromatic tar

compounds in the gas increases significantly. Staging of the gasification agent, oxygen and steam, despite being potentially interesting, has not been reported. It seems that a more distinct division of zones in the gasifier is necessary for further tar reduction; however this makes it necessary to leave the conventional design as we will see in Section 4.

The presence of an in-bed catalyst and local gas and solids mixing has a great influence on the cracking and/or reforming reactions [6,9-11]. Typical in-bed catalysts are calcined limestone, dolomite, and olivine sand and, less frequently, Ni-based or other metallic catalysts. Reduction of tar by cheap catalysts, such as natural mineral (dolomite, olivine, etc.) reaches tar concentrations of down to 0.5 g/Nm^3. However, the remaining small amounts of heavy tar lead to a gas with a dew point around 100 °C and this will cause problems in applications where the gas has to be cooled to the ambient temperature, so this method has to be complemented by secondary treatment. More effective catalysts, such as those based on nickel, improve tar reduction in the bed, but the rapid degradation of the catalyst makes this option unfeasible. Ni-based catalysts are preferred in downstream vessels, but deactivation is a problem still to be solved [12]. Commercial activated carbon has also been found to be an active catalyst of tar [13,33-36]. Char generated in the process acts as a catalyst due to the alkali and alkaline earth-metals remaining in its structure, especially potassium in the form of carbonates, hydroxides or oxides, which has an effect on steam reforming of the nascent tar [34,35]. Char undergoes significant transformation during the conversion process and it is simultaneously gasified by steam in the fluidization gas. Polimerization of PAH tar compounds at temperatures above 700°C [36] lowers the rate of char gasification with steam, whereas below 600°C the char acts as a tar filter reducing significantly the tar concentration although it is not gasified.
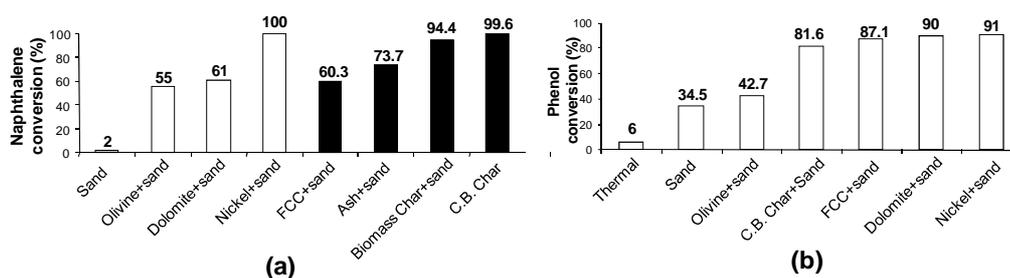

Figure 7: Effect of various catalysts on (a) naphthalene conversion at T=900°C and τ=0.3 s; and (b) phenol conversion at T=700°C and τ=0.3 s; (the colour of the bars indicate different initial tar concentration. (Adapted from El Rub et al. [33]).

To give a rough idea on the effectiveness of distinct catalysts, experimental findings [33] have been illustrated in **Figure 7**. The measurements comprised two model compounds: phenol and naphthalene. The figure shows a ranking of the activities of catalysts (and also thermal cracking) for conversion of naphthalene at 900 °C (a) and for phenol at 700°C (b). Naphthalene is thermally stable at 900°C (only 2% was converted over silica sand), whereas it is more converted with dolomite and olivine, and almost fully converted in the presence of nickel-based catalyst and char. It is seen that the results are quite insensitive to the tar concentration in the inlet gas (the inlet concentration is that typically found in FBG). The representation for phenol conversion is made at 700°C because at 900 °C all catalysts gave 100 wt% phenol conversion, and more than 98 wt.% of the phenol was already thermally cracked. Although the figures are obtained in specific conditions [33], the results lead to important conclusions: the conversion of phenol is not a problem in FBG at temperatures over 800-850°C, whereas the reduction of naphthalene, or a mixture of tar compounds with similar reactivity, down to 0.5–1 mg, is difficult in an FBG. Char effectively converts the heavy tar compounds, so optimization of the contact of tar with in-situ generated char might be targeted. However, this it is difficult to achieve in a single vessel owing to fluid-dynamic effects, as shown in the previous sections. Staged gasification could make this concept possible, as we will see below.

As a main conclusion, the combined use of in-bed catalysts, injection of secondary air and optimization of the composition of the gasification agent in conventional FBG designs, despite improving the process, were shown to be insufficient to attain the gas purity required for cold gas applications. It is concluded that further measures involving redesign of the gasifier/process are necessary, i.e. to develop innovative designs. One rational option to do this is dealt with in the following section.

Innovative designs based on stage gasification

It has been shown that optimization of conventional directly heated FBG is not effective enough for tar reduction (dew point). Indirectly heated gasifiers consisting of twin beds convert most of the char, but the tar problem remains, so it is not a sufficiently good solution as long as expensive secondary cleaning of the gas is needed. Therefore, staged gasification in directly heated gasifiers has been proposed, searching for high conversion of both tar and char in the reactor itself [37–46].

Staged gasification creates zones in the gasifier, allowing to optimize the process by increasing simultaneously the conversion of char and tar. The essential idea is similar to that conceptualized in Figure 8(a) but here a more drastic zone division than that of secondary air injection is achieved by dedicated design. Various thermal levels are created in the gasifier by staging the oxidant: the fuel is devolatilized at relatively low temperature by decreasing the amount oxygen to this zone; the temperature is increased in a second zone by addition of the remaining oxygen together with steam. This two-stage procedure favors the conversion of tar because it creates a gas with high-reactive tar compounds at high temperature in the presence of steam (non-catalytic gas-phase reforming). If the char generated is made to form a third stage (see in Fig. 9(b)) the char can be gasified with steam and further tar can be converted. A example of the three-stage concept is shown in Figure 8(b). In this figure devolatilization is conducted without addition of oxygen [41]. The heat for pyrolysis comes from that generated in the second stage, conducted by successive heat exchanges (not shown in the figure).
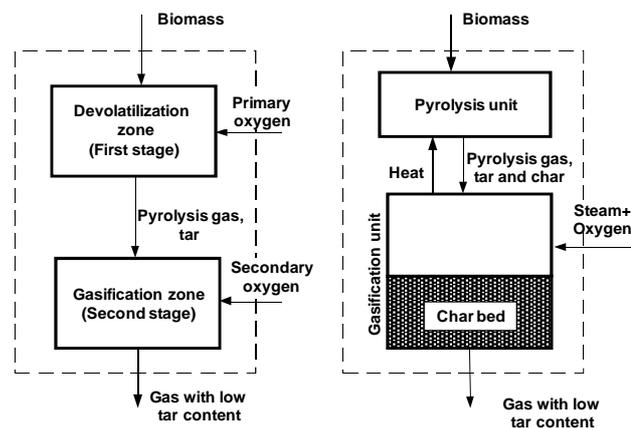

Figure 8: Staging of the gasification process: (a) secondary air injection. (b) Two-stage gasifier

A few innovative processes have been proposed based on staged gasification. Examples are processes like CASST, developed at Energy research Centre of the Netherlands [42], the TSG at Danish Technical University in fixed bed [41] and fluidized bed [43], STAR-MEET at Tokyo Institute of Technology [37], CleanStgGas at ITE Graz University of Technology [44], and other [45,46]. In most of this type of gasifiers the char is converted by gasification (with steam or CO_2), so the efficiency of the process depends on how the conversion is "organized". Since

char gasification reactions are slow, it is necessary to provide long residence time to achieve significant char conversion. Therefore, most such processes have been based on fixed bed designs where this easier can be achieved. A process combining fluidized and moving beds [39,46] has been suggested recently, oriented to the conversion of difficult waste, with high fuel utilization but the tar content in the gas is still high.

A new three-stage gasification system for biomass and waste (FLETGAS)

All staged gasification designs mentioned where high conversion of tar and char has been achieved are fixed or moving beds. In order to carry out staged gasification, enabling high throughputs, a new staged-gasification concept based on FB is under development by the Bioenergy Group at the University of Seville [47]. The system is focused on processing of difficult wastes, whose ash content is high. The nature of the ash limits the temperature of the gasifier because of the risk for agglomeration.

The gasification concept is based on three stages: FB devolatilization (first stage), non-catalytic air/steam reforming of the gas coming from the devolatilizer (second stage), and chemical filtering of gas in a moving bed supplied with the char generated in the devolatilizer (third stage). The operational principle of the system is outlined in Figure 9, where the direction of the flows of solids (biomass, bed material and char) and gas (inlet flow rates of fluidization agent) are indicated. A control valve is included to adjust the residence time of the solids through the control of the solids inventory in the loop.
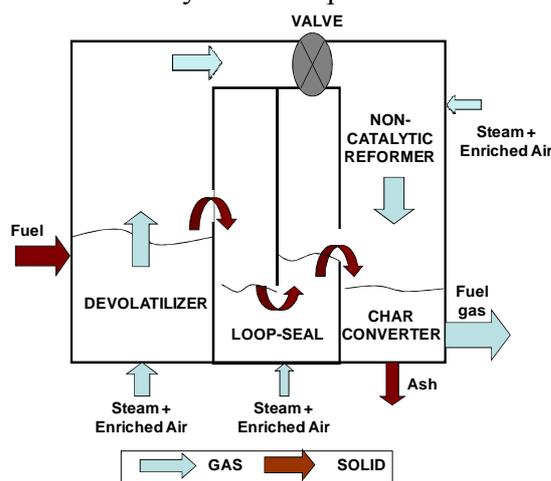

Figure 9: Conceptual performance diagram of FLETGAS process.

Air and steam can be injected at various points (in the devolatilizer, steam reformer and seal) with different proportions of the two reactants. Enriched air, with an oxygen concentration of up to 40% (to keep the price reasonably low, for instance, produced by membranes) can be used instead of air. The fuel is fed near the bed's surface and has to circulate down to the bottom before leaving the bed. The devolatilizer, where a high yield of fresh tar is generated, is operated at relatively low temperature (700-750°C). The fresh tar compounds are drastically reduced in the reformer where a temperature of up to 1200°C is created. The injection of steam into the reformer avoids coking and polymerization of tar. The gas is filtered in a moving bed made of char coming from the loop seal. The loop seal can be operated as an oxidiser (fed with enriched air) or as a light reformer (fed with H_2O) depending on the fuel's reactivity and ash properties. The char filter also cools down the gas (chemical quench) by the endothermic char gasification reaction with steam, while it acts as a catalytic filter promoting tar decomposition reactions with steam.

It is challenging to achieve the appropriate movement of solids and gas, so a cold model (scaled-down from an imaginary 2MW$_e$ gasification plant, operating with dried sewage sludge)

was built [47]. A fluid-dynamic model was developed to understand the movement of gas and solids, predicting the distribution of mass of solids between the bed and loop seal [76]. Fuel conversion tests (devolatilization, char gasification, and tar conversion) in a bench-scale FBG have also been conducted [48]. The data from the cold and fluid-dynamic models and from the fuel and tar conversion tests have supported the design of a plant that is currently under construction to demonstrate this new gasification concept at pilot scale (30 kg/h).

Conclusions

Measures to optimize fuel conversion in an FBG to simultaneously achieve high tar and char conversion have been reviewed. Treatment of tarry gas downstream of the gasifier is complex and expensive for power production in small/medium size gasification plants, so methods to improve tar conversion within the gasifier are preferred. The optimization of conventional FBG by in-bed catalysts and distribution of the gasification agent, although improving the process, is shown to be insufficient to attain the gas purity required for such an application. Staged gasification creates zones in the gasifier, which promote high conversion of char and tar and, therefore, it is an effective and feasible way to conduct gasification. Various developments have been proposed (based on fixed bed) for small-scale gasification over the last decade, but none of them has still reached commercial status. A new three-stage gasifier (FLETGAS) based on fluid-bed design has been presented here. The new process is promising because it enables to increase the flexibility and capacity of existing staged gasification developments. The FLETGAS process is still under development at pilot scale.

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