



Analysis of gasification and tar reforming with in-situ generated char and steam
Análisis de la gasificación y reformado de alquitrán con carbón y vapor generado in-situ

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Abstract

Biomass gasification has an overwhelming potential to satisfy the world's energy needs. Nevertheless, the use of this technology is constrained by the formation of tars and the costs associated to their eradication. Catalytic steam reforming is a common approach for their removal but requires the addition of steam and a catalyst. At the same time, char can be used as a catalyst and, similarly to steam, can be produced during gasification. This work presents an analysis, based on numerical simulations, to elaborate a concept where the steam and char released from gasification are used to reform the produced tars. The results indicate that when the humidity in the biomass (wood in this study) is 8 wt% or higher, at adequate temperatures (around 1073K) and with a residence time of 0.5 seconds, both the steam volume and the char produced meet the necessary criteria for char catalytic tar reforming during each gasification run. Moreover, it was found that moisture in biomass leads to increases in the Lower Heating Value of the produced syngas from 5.6 to 5.9 MJ/Nm³, and decreases in the tar contents, from 56 to 19 g/Nm³. Based on theoretical calculations, the concept appears promising and should be subject to experimentation.

Keywords: Biomass gasification, tar reforming, auto-generated steam, syngas, char catalyst

Resumen

La gasificación de biomasa tiene un enorme potencial para satisfacer las necesidades energéticas del planeta. Sin embargo, la tecnología está limitada por la formación de alquitranes y los costos asociados a su erradicación. El reformado catalítico es una opción comúnmente utilizada para su remoción, pero requiere la adición de vapor de agua y un catalizador. Por otro lado, el carbón puede ser usado como catalizador y, de forma similar al vapor de agua, puede ser producido regularmente durante la gasificación. Éste trabajo presenta un análisis basado en simulaciones numéricas para proponer un concepto donde el vapor y carbón liberados durante la gasificación se usan para reformar los alquitranes. Los resultados indican que cuando la humedad en la biomasa (madera en este estudio) es 8% en peso o superior, a temperaturas adecuadas (alrededor de 1073K) y con un tiempo de residencia de 0.5 segundos, tanto el volumen del vapor como carbón producidos cumplen los criterios necesarios para garantizar el reformado de los alquitranes. Además, la humedad en la biomasa incrementó el Poder Calorífico Inferior del syngas producido de 5.6 a 5.9 MJ/Nm³, y redujo la concentración de alquitranes 56 a 19 g/Nm³. Según los cálculos teóricos, el concepto es prometedor y debe ser sujeto a experimentación.

Palabras clave: gasificación de biomasa, reformado de alquitrán, vapor autogenerado, syngas, catalizador de carbón.

1 Introduction

The concerns for global warming have increased, along with a search for the mitigation of the environmental impacts associated to fossil fuels, turning the use renewable energy from a luxury to a necessity. Although renewable energy can be obtained from different sources such as the sun and

the wind, biomass is of special interest because of its non-intermittence and virtually inexhaustible nature (Brown, 2011; Basu *et al.*, 2013). The energy found in biomass can be harnessed via different thermal conversion technologies such as pyrolysis, torrefaction and gasification. During pyrolysis, the biomass is heated under an inert atmosphere, leading to the degradations of the hydrocarbons in the biomass into smaller molecules, whose mixture is often termed pyrolysis oil (Brown, 2011).

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Table 1. Reactions in biomass gasification that involve steam.

Steam gasification	$C_{(s)} + H_2O \rightarrow H_2 + CO$	(1)
Steam reforming (1)	$C_nH_m + 2nH_2O \rightarrow \left(\frac{m}{2} + 2n\right)H_2 + nCO_2$	(2)
Steam reforming (2)	$C_nH_m + nH_2O \rightarrow \left(\frac{m}{2} + n\right)H_2 + nCO$	(3)
Water-gas shift	$H_2O + CO \rightarrow H_2 + CO_2$	(4)

Torrefaction is a similar process, but the operation pressure is much higher (up to around 10 times), and the results end up being carbonaceous solid fuels (Brown, 2011). Finally, during gasification, the oxygen supply is restricted to prevent complete combustion of the biomass and a mixture of gases called syngas is produced (Brown, 2011). Although the process can vary, generally, gasification is comprised of several stages, all of which normally occur in the same reactor: drying, pyrolysis, oxidation, and reduction (Sikarwar *et al.*, 2016). During drying, the biomass loses all the moisture. Following, the pyrolysis stage occurs, where the biomass is decomposed into hydrocarbons (including aliphatic and aromatic type) at a temperature between 500 and 700K, depending on the process. The oxidation stage occurs next, where the gasification agent (air, steam, oxygen or sometimes CO₂) reacts with the hydrocarbons at temperatures up to 1400K to form the gasification products: H₂, CO, CO₂ (the syngas) and other trace components, such as a hard to decompose mixture of hydrocarbons and oxygenated compounds called tars (Sikarwar *et al.*, 2016; Woolcock *et al.*, 2013). The final stage of gasification (reduction) aims to degrade the remaining hydrocarbons into syngas and occurs at a wide interval of temperatures, between 900 and 1200K.

Gasification has attracted the interest of scientists not only because the produced gas can be combusted for energy, transformed into liquid fuels or upgraded into high value fuels, amongst other uses, but because the process is more efficient than other similar technologies such as pyrolysis (Basu *et al.*, 2013; Woolcock *et al.*, 2013; Zhang *et al.*, 2016). Nonetheless, gasification has some challenges, such as the removal of the tars. Tars condense at relatively high temperatures (more than 400K) and need to be removed for most syngas applications Basu *et al.*, (2013). Research efforts have led to the conclusion that catalytic reforming is the most plausible way of eradicating the tar while at the same time producing hydrogen (Woolcock *et al.*, 2013; El-Rub *et al.*, 2008).

However, catalysts often suffer from deactivation by tar coke deposition and thermal stability failures, and are therefore limited by the need for constant replacement. Although the catalysts that provide the most tar conversion (around 100% at temperatures circa 1000K) are transition metal-based (i.e. use Ni, Zn, Fe, etc) (Woolcock *et al.*, 2013), char has proven to be a reliable catalyst as well. Char offers more than 90% conversion at moderate temperatures (circa 1123K), resists coking under a steam atmosphere and at adequately high temperatures (around 1100K) and as a catalyst, is “auto-generated” during gasification (El-Rub *et al.*, 2008; Buentello-Montoya *et al.*, 2020; Fuentes-Cano *et al.*, 2013).

On the other hand, some authors have reported interactions between the steam generated during pyrolysis of biomass and char particles in a reactor, enhancing the production of hydrogen and CO rich fuel gas (Sun *et al.*, 2011; Hayashi *et al.*, 2000; Xiong *et al.*, 2013; Al-Rahbi *et al.*, 2017). The main reactions taking place during gasification and involving steam can be found in Table 1.

Fuentes-Cano *et al.* (2013) observed that the influence of steam on the conversion of tar was only significant in a very particular range of steam concentration and temperature, from 0% to 15% at 1223 K. Steam participates in reforming reactions and is necessary for them to occur (as found in Eqns. 2 and 3, in Table 1), but studies have shown that its concentration has a marginal influence in the reaction rate, in agreement with the zeroth order of the reactions with respect to steam reported in literature (Fuentes-Cano *et al.*, 2013; Świerczyński *et al.*, 2007; Phuphuakrat, 2010; Jess, 1996). Previous studies tested using char with steam concentrations between 0 and 25% volume and found that as long as there is at least some steam in the gas mixture, the reaction rate was not influenced by the steam concentration (Fuentes-Cano *et al.*, 2013; Świerczyński *et al.*, 2007; Phuphuakrat, 2010). Water has a relatively high heat capacity (4.187 kJ kg⁻¹ K⁻¹) and some energy is necessary for its evaporation during drying, thus large

amounts of moisture are unfavourable and unwanted in any biomass to be gasified. For a biomass moisture content of 66 wt.%, the whole product gas has to be used to maintain the gasification process, bringing the efficiency down to zero (Schuster *et al.*, 2001). However, should the moisture content be adequate, the steam generated when the biomass is dried could be recovered and used for reforming, mitigating the energetic penalty associated to steam generation for reforming; as reference, around 2260 kJ are necessary to evaporate one kg of steam from ambient temperature (Brown, 2011; Dahiya, 2014). Auto-generated steam, together with auto-generated char, could increase the sustainability of the gasification process, reducing potential additional costs of steam generation and acquiring commercial catalysts. This is an area of opportunity, as under adequate conditions, the steam produced during the drying and oxidation stages of gasification may suffice for reforming reactions to occur. To date, no dedicated studies involving the gasification of wet biomass, and only a few studies involving pyrolysis of wet biomass (Hayashi *et al.*, 2000; Luo *et al.*, 2017) were found. No studies have been dedicated to the use of auto-generated steam as a gasification tar reforming agent.

At the same time, staged gasification and catalytic reforming either in a single reactor or in a sequence is an emerging approach pursued worldwide due to its efficiency (Sikarwar *et al.*, 2016; Corella *et al.*, 1999). During staged gasification, the gasification “stages” (drying, pyrolysis, oxidation, etc.) take place separately and sometimes unconventionally. Staged gasification has seen a growth in popularity because of the low tar yields ($< 500 \text{ mg/Nm}^3$) and high carbon conversions ($> 90\%$) obtained, and has been researched worldwide to different results (Leijenhorst *et al.*, 2015; Henriksen *et al.*, 2006; Zeng *et al.*, 2016; Wang *et al.*, 2007; Trippe *et al.*, 2011). For example, the group led by Henriksen in the Technical University of Denmark (Henriksen *et al.*, 2006; Gadsbøl *et al.*, 2019) developed a pioneering staged gasifier system, where in contrast with conventional gasification, pyrolysis and char gasification occur in separate reactors. Other staged gasifiers have secondary air injection in a separate reactor. For example, the Biomass Technology Group in the Netherlands (Leijenhorst *et al.*, 2015) proposed the use of low temperature staged gasification using Ni and Pt-based catalysts, using an air-vapor mixing device with air entering the vapour steam through many small holes and three additional mixing devices to increase turbulence and found that increasing the

turbulence during gasification resulted in changes in the hydrocarbons composing the tar mixture. Zeng *et al.* (2016) proposed a design for an autothermal two-stage fluidised biomass gasification reactor; while the tar yields obtained from the experiments are still relatively high (around 400 mg/Nm^3), the proposed reactor has a relatively high capacity (600 kg/h) for a novel technology. Based on emerging approaches, the overall aim of this work is to numerically simulate and present a concept for the development of a novel staged-gasification system that uses a fluidised bed of char and steam generated in the gasifier itself.

The first part of the manuscript presents the methodology used to do the necessary calculations and biomass gasification simulations to support the proposed theoretical concept. Afterwards, the novel gasification concept is described, along with calculations to estimate the required char and steam. The following sections describe the results and analysis from the calculations. In more detail, subsections elaborate on the effect of moisture content during gasification, the effect in the amount of steam produced, the amount of char produced under different sets of conditions and how steam and char could synergically interact to increase the efficiency of gasification by providing “free” catalysts/reagents. Subsequently, recommendations to successfully employ the novel concept are given. Finally, a summary and conclusions of the overall findings are given in the closing section.

The physical design, construction and testing of the proposed gasifier, however, remain beyond the scope of this writing. Moreover, there are several studies that document the reforming of tar using char (Buentello-Montoya *et al.*, 2020; Fuentes-Cano *et al.*, 2013; Burhenne *et al.*, 2014), hence, the kinetic analysis behind tar reforming is not covered in this work.

2 Materials and methods

2.1 Overview of the numerical model

The calculations of the gasification process were conducted based on a stoichiometric thermodynamic equilibrium approach (i.e. steady state has been achieved). Previous studies have indicated that a thermodynamic equilibrium approach is suitable to simulate downdraft and fluidised bed gasifiers (Lim *et al.*, 2014, Schuster *et al.*, 2001). To simulate the

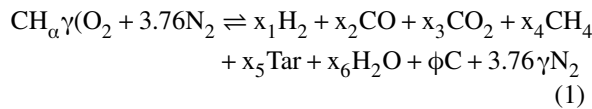
equilibrium composition of the syngas and solids produced, a number of assumptions were necessary, namely:

- The gasified biomass was that from Tuomi *et al.* (2015) and is composed, in % wt, by C: 53.2%, H: 5.5%, O: 37.1%, N: 0.3%, Ash: 3.9%.
- The produced gases behaved as ideal gases.
- The tar was considered to be composed by three popularly employed model compounds: benzene, toluene and naphthalene (Fuentes-Cano *et al.*, 2013; Jess, 1996; Taralas et al, 2003).
- The air used for gasification is assumed to be at the gasification temperature. This is adequate under thermodynamic equilibrium, as the partial oxidation reactions involved are exothermic in nature and lead to increases in temperature.

Different temperatures (923 - 1173 K), equivalence ratios (0.25 - 0.45) and biomass moisture contents (0 - 8 % weight) were used during the simulations.

2.1.1 Mass balance

The concentrations of the biomass gasification products were obtained from a global gasification reaction that considers biomass and air as reactants and H₂, CO₂, CO, CH₄, steam, char, N₂ and tar as products (Brown, 2011):



where α and β correspond to the stoichiometric relationship (from weight percentages obtained by the ultimate analysis of biomass), γ is the molar quantity of air introduced to the gasifier, x_i are the molar quantities of products from gasification and ϕ is the molar amount of unconverted char, calculated from the relationship proposed by Lim *et al.* (2014):

$$\phi = 0.901 + 0.439(1 - r^{ER+0.0003T_{gas}}) \quad (2)$$

where ER is the equivalence ratio in the gasifier and T_{gas} is the gasification temperature, in K.

The equivalence ratio is defined as the actual ratio of fuel to air divided by the stoichiometric fuel to air ratio. In other words,

$$ER = \frac{m_{fuel}/m_{air}}{(m_{fuel}/m_{air})_{st}} \quad (3)$$

where m_{fuel} is the fuel mass, m_{air} is the air mass and st stands for stoichiometric.

To complement the mass balance equation for the simulations, equilibrium equations for methanation, water-gas shift and carbon gasification were used (Font Palma, 2013).

2.1.2 Energy balance

The energy balance for the gasification reaction was defined as (Smith, 1950):

$$\Delta H_{biomass}^0 + \gamma(\Delta H_{O_2} + 3.76\Delta H_{N_2}) = \sum x_i \Delta H_i + Q_{lost} \quad (4)$$

where ΔH_i are the changes in enthalpy of species i , $H_{biomass}^0$ is the heat of formation of the biomass at 298 K and Q_{lost} is the heat lost through reactor walls by diffusion. The changes in enthalpy for species i were defined as (Smith, 1950):

$$\Delta H_i = \Delta H_i^0 + C_{p,i}(T_{gas} - T_{atm}) \quad (5)$$

where $C_{p,i}$ is the specific heat at constant pressure for species i and T_{atm} is the ambient temperature (298.15 K). ΔH_i^0 of several compounds has been widely documented and can be found in textbooks (Smith, 1950). On the other hand, the heat of formation for the biomass, $\Delta H_{biomass}^0$, depends on the biomass composition and needs to be calculated following the relationship (Lim et al, 2014):

$$\Delta H_{biomass}^0 = \Delta H_{CO_2}^0 + \frac{\alpha}{2} \Delta H_{H_2O}^0 + (12 + \alpha + 16\beta) LHV_{biomass} \quad (6)$$

$$LHV_{biomass} = 34.835w_C + 93.87w_H - 10.8w_O + 6.28w_N + 10.465w_S \quad (7)$$

where $LHV_{biomass}$ is the biomass Lower Heating Value (MJ/kg), w_C , w_H , w_O , w_N and w_S are the mass fractions of carbon, hydrogen, oxygen, nitrogen and sulfur (ignored in this work) in the biomass respectively.

2.2 Concept proposal

The biggest problems for gasification are associated to the tar clean-up stage, the energy consumed during the process and the cost of generating the gasification agent (Abdoulmoumine *et al.*, 2015; Roberts *et al.*, 2007). A practical system should be inexpensive and simple while providing the desired products.

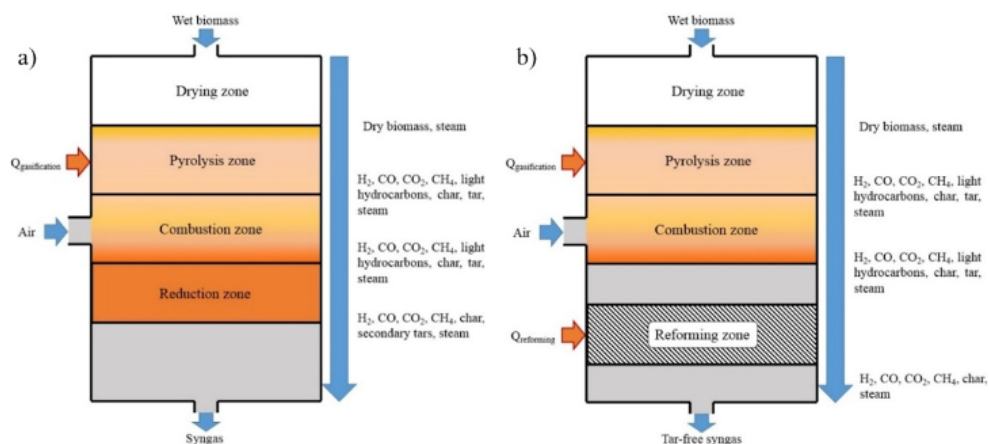


Fig. 1. Scheme outlining a) a conventional downdraft gasifier and b) the theoretical two-stage reactor proposed in this work.

This could be done by recovering some of the residuals from gasification and recirculating them into the system: char and steam.

Although air as a gasification agent provides a syngas with a lesser Lower Heating Value when compared to oxygen and steam, it is cheap and reliable, and allows for autothermal gasification (Brown, 2011). For that reason, air is used as the gasifying agent in this work.

With regards to reactor configuration, fluidized bed gasifiers offer good carbon conversion but are generally more complicated (and expensive) than fixed bed gasifiers, and can have sintering problems at temperatures higher than 1073 K (Sikarwar *et al.*, 2016), thus a simple fixed-bed reactor was chosen as a baseline for the concept.

A simplified scheme, where the proposed system is compared with a conventional downdraft gasifier, is found in Figure 1. In the proposed system (Figure 1b), the first gasification stage comprises the conventional drying, pyrolysis and oxidation stage, while the second stage consists on reforming with a fluidised char bed. The second stage, which should be positioned downstream from the oxidation zone in the reforming zone, uses auto-generated steam (i.e. generated from the biomass moisture) for reforming and a bed of char from the gasification process as a catalyst. This setup could lead to improvements in the process efficiency by reducing the penalty associated to external steam generation.

2.2.1 Char recovery for catalysis

In order to determine the feasibility of using the char produced during gasification as a catalyst, the theoretical amount of char needed in a fixed bed for a certain gas yield was calculated. The required char mass was calculated as a function of the gas yield from the bulk density of char and the gas residence time in the char bed using the following equations (Cengel *et al.*, 2006; Levenspiel, 1999):

$$t = \frac{V_{reactor}}{Q} \quad (8)$$

$$\rho_{char} = \frac{m_{char}}{V_{char}} \quad (9)$$

where t is residence time, $V_{reactor}$ is the bed volume, Q is gas flow rate, ρ_{char} is the bulk density of the char, m_{char} is the mass of char and V_{char} is the volume of char. A value of 103 kg/m³, calculated by weighting the compressed mass used in the reactor bed to conduct previous experiments (Buentello-Montoya *et al.*, 2020), was used as the bulk density for the calculations. The syngas flow rate is influenced by the reaction conditions i.e. temperature, thus, the effect of the temperature was incorporated using the ideal gas law:

$$PV = nRT \quad (10)$$

where P is the gas pressure in Pa, V is the gas volume in Nm³ (or volumetric flow rate in an open system), R is the ideal gas constant (Pa·m³·K⁻¹·gmol⁻¹) and T is the temperature in K.

2.2.2 Usage of auto-generated steam for tar reforming

Heating water to achieve the temperatures required for reforming consumes a large amount of energy (at least 2271 kJ/kg to evaporate at 100 °C (Lim et al, 2014)). An estimate can be obtained by observing the widely documented steam tables for saturated water vapour (Smith, 1950). From the steam tables, the necessary energy to superheat water from a saturated liquid at 298 K to 1173 K (necessary for complete tar reforming (Buentello-Montoya et al, 2020) and 1 atm is 4.396 MJ/kg_{steam}.

If the gasifier is assumed to be at thermodynamic equilibrium, the steam generated from biomass drying should be at the gasification temperature. Therefore, the necessary energy to increase the temperature from gasification can be calculated from energy balances, following the equation (Smith, 1950):

$$\Delta H_{ref} = \sum_i C_{p,i}(T_{ref} - T_{gas}) \quad (11)$$

where ΔH_{ref} is the energy needed to achieve a reforming temperature from gasification temperature, $C_{p,i}$ is the specific heat of species i , T_{ref} is the reforming temperature and T_{gas} is the gasification temperature.

3 Results and discussion

3.1 Effect of moisture in the gasification of biomass

To determine the effect of the moisture contents (MC) in the reference biomass, several simulations were conducted using in-house developed code written in Python 3.0. The syngas LHV and tar contents were determined at different temperatures, equivalence ratios and moisture contents. Figure 2 shows the effect of temperature and ER at different MCs.

Figure 2 shows that increasing the moisture content increases the LHV and decreases the tar concentration, regardless of the equivalence ratio. The LHV increased by around 0.2 MJ when the moisture content increased from 0 to 8 wt.% and gained an additional 0.1 MJ in LHV when the moisture content was increased again, from 8 to 16 wt.%. It can be seen that the steam content had a drastic impact in the tar concentration, particularly at 973 K, where the tar concentration decreased by around 50% when the steam concentration was increased from 0 to 16 wt.%. These results agree with the observations reported by Gao et al. (2014). As expected, both the syngas LHV and tar contents decrease with increasing equivalence ratio.

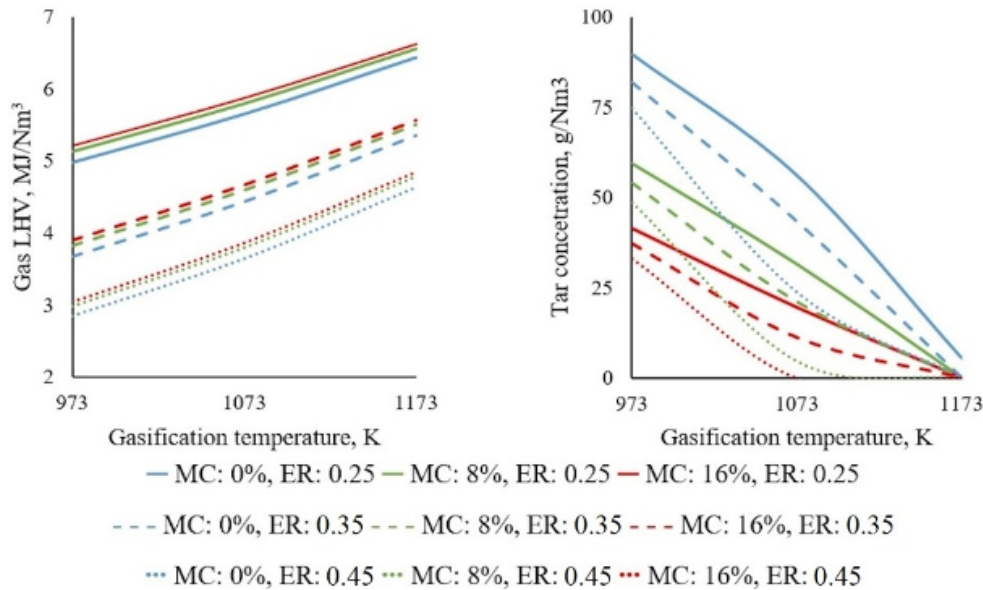


Fig. 2. Effect of temperature in the syngas (a) LHV and (b) tar contents from gasification of biomass with different moisture contents.

On the other hand, the LHV increases with increasing temperature, while the tar contents decrease. In Figure 2b it can be seen that at high gasification temperatures of 1173 K, the tar contents are close to zero, making the use of a reforming reactor appear unnecessary. However, since char-based catalysts promote the water-gas shift reaction, and depending on the end use of the gas, the inclusion of a reforming stage is more beneficial to the overall process than conducting gasification at high temperatures (Abdoulmoumine *et al.*, 2015; Buentello-Montoya *et al.*, 2019).

3.1.1 Use of auto-generated steam for tar reforming

Fuentes-Cano *et al.* (2013) conducted gasification experiments varying the steam concentration during tar reforming over char, and reported that when the steam contents are around 15% volume, the coke deposited in the char (regardless of their porous structure) can be gasified at 950 °C. (Buentello-Montoya *et al.*, 2020). In order to know whether the steam produced from biomass gasification was

enough for reforming, simulations were conducted using in-house developed code, written in Python 3.0, to determine the equilibrium concentrations of steam after gasification as a function of the biomass moisture content, as found in Figure 3. The effect of the moisture content is more evident at low gasification temperatures and equivalence ratios. The decrease in steam volume from 16% to 5%, with increasing temperature from 973 K to 1173 K occurs due to the water-gas shift reaction (Eq. 4), which leads to the formation of H₂ from steam. For a gasification temperature of 1073 K, the steam content remained at values around 10%. Based on literature results (Fuentes-Cano *et al.*, 2013), this might be sufficient for the system to work continuously with a constant reforming activity. However, focused experiments need to be conducted to ascertain whether this amount is enough for continuous coke gasification to occur. Worth mentioning is that, even when the moisture content in the biomass is 0%, some steam is produced during the partial oxidation of the biomass. The steam contents in the syngas mixture decreases with increasing equivalence ratio.

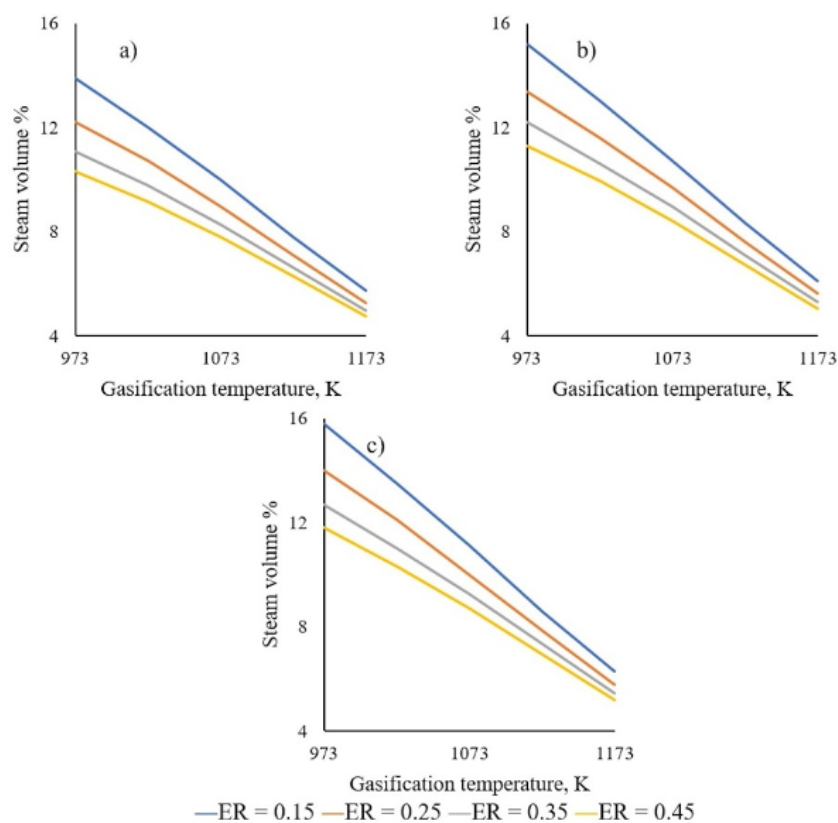


Fig. 3. Volume of steam in the syngas produced from biomass gasification as a function of temperature and equivalence ratio, when the moisture content in biomass is (a) 0%, (b) 8% and (c) 16%.

Solids moisture have been proven to be prejudicial to the gas quality. Xiong *et al.* (2013) found that when the moisture content is more than 50% wt., the excess steam leads to the reactor cooling down and decreases in efficiency and syngas quality.

3.1.2 Comparison of energetic requirements: external vs auto-generated steam

Since gasifying at different equivalence ratios results in different gas yields (stemming from different ER) (Zhang, 2016), the energy necessary to increase the temperature from a certain gasification temperature T_{gas} to a reforming temperature of 1173 K (to achieve complete tar conversion) was calculated at different equivalence ratios and is found in Figure 4.

Clearly, lower gasification temperatures and equivalence ratios require larger energy inputs. This is associated to the temperature differences and the syngas composition; for example, at a gasification temperature of 923 K the difference of energy consumption between gasifying at an ER of 0.15 and an ER of 0.45 is around 0.05 MJ/kg, and becomes negligible at a gasification temperature of 1123 K. As informative calculations, an analysis of the energy consumption for gasifying the biomass from a reference case follows. When gasification is done at 1073 K and an equivalence ratio of 0.25, the LHV of the produced syngas is 5.96 MJ/Nm³, and the gas yield is 2.3 Nm³/kg_{biomass}. This means that, per kg of biomass, around 13 MJ could be produced. Considering that the amount of steam used for reforming is 10% of the total gas yield, the total energy consumption to increase the temperature of steam from ambient to a temperature of 1173 K, calculated by using the values from the steam table is at least 0.96 MJ, which is around 16% of the energy produced.

In contrast, the heat necessary to increase the temperature from gasification temperature to a reforming temperature of 1173 K is less: From a gasification temperature of 1073 K using an equivalence ratio of 0.25, around 0.06 MJ are necessary when considering the staged gasifier with auto-generated steam setup. This is around 7% of the energy that would be necessary considering the injection of external steam. Although the

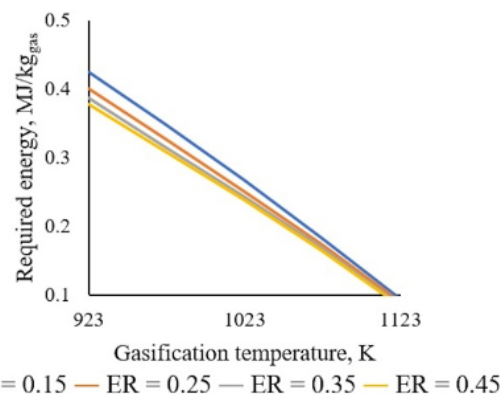


Fig. 4. Energy required to increase the temperature of the syngas in a two-stage reactor, from an initial gasification temperature to a final reforming temperature, as a function of gasification temperature and Equivalence Ratio.

energy savings appear to scale up with the gas yield, the calculations are theoretical and actual experiments are necessary to determine the operability of the system.

3.2 Recovery of char for use as a catalyst

Figures 5a - 5c show the amount of char required to achieve certain residence time of the gas produced from gasification in a char-bed at (a) 650, (b) 750 and (c) 850 °C, while Figure 5d shows the amount of chars produced from gasification at different equivalence ratios. No differences are seen between the necessary carbon at temperatures shown in Figures 5a-c. Contrastingly and expectedly, higher gas yields require more char to achieve the desired residence time. References (Fuentes-Cano *et al.*, 2013; Burhenne *et al.*, 2014) have indicated that, when the residence time in a char bed is around 0.5 seconds, complete tar reforming can be achieved at reforming temperatures around 1173 K. For that residence time, more than double the required mass was obtained for all the values of equivalence ratios and gas yield. Importantly, the char may undergo deactivation during reforming and may require replacement. However, from the calculations and in every set of conditions, the char produced is sufficient to replace the deactivated char and ensure constant tar reforming. This corresponds to theoretical estimations, and proper experimental work will be featured in future communications.

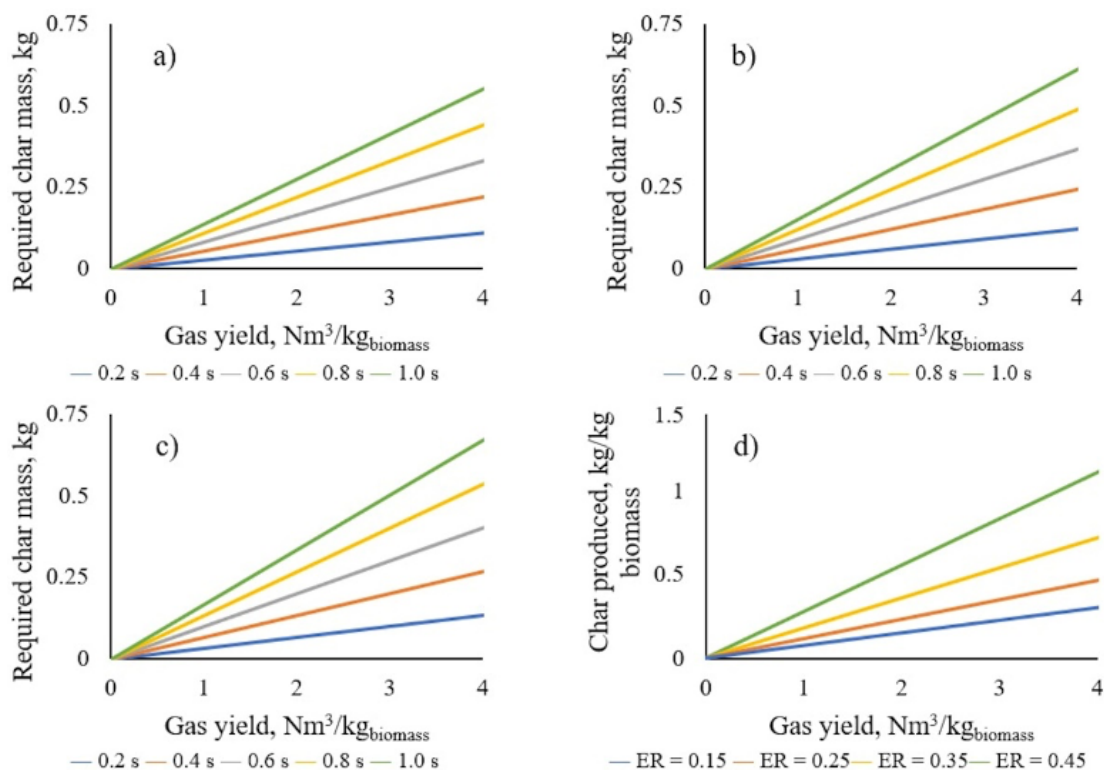


Fig. 5. Char mass required to achieve certain residence time as a function of gas yield at different temperatures. Figures a - c correspond to the required char mass to achieve a specific residence time at (a) 650 °C, (b) 750 and (c) 850 °C, while figure (d) represents the char produced per kg biomass at different equivalence ratios.

3.3 Possible applications of the concept

Generally speaking, downdraft gasifiers do not operate adequately when the biomass moisture content is more than 25% wt. (Pereira *et al.*, 2012). However, the hydrophobic quality of a gasifier is a problem not only for downdraft gasifiers, but for gasifiers in general (Pereira *et al.*, 2012; Bridgwater *et al.*, 2002). Notwithstanding, Section 3.2 showed that it is theoretically possible to save energy by using steam from the biomass moisture instead of externally produced steam. Based on the calculations from Section 3.2, a set of different biomasses were studied and chosen as “suitable” for application of the concept. Table 2 shows the average ultimate and proximate analysis of said biomasses, retrieved from Vassilev *et al.* (2010). Based on their moisture and ash contents, straw and most woody biomass are potential feedstocks. Even if downdraft gasifiers generally have

low tar yields, the tars formed during gasifier are heavier and more refractory (i.e. more stable) in nature than those from other gasifiers, and if the desired use is for internal combustion engines, the tar restrictive allowable tar limits establish a need for clean-up (Morf *et al.*, 2002). However, since naphthalene, one of the most stable of the heavy tars, has been successfully reformed at temperatures of around 1123 K using char as a catalysts, it is safe to assume that most if not all tars can be reformed with the proposed design (Fuentes-Cano *et al.*, 2013). Therefore, even if the tars are more stable than those formed in the other kinds of gasifiers, catalytic reforming downstream of a downdraft gasifier should ensure satisfactory tar clean-up, resulting in syngas with enough cleanliness for most applications, for example, if used in internal-combustion engines, tar concentration in the product gas should be less than 100 mg/Nm³ (Basu *et al.*, 2013).

Table 2. Average, minimum and maximum values of the ultimate analysis composition of some biomasses, in weight%. Adapted from Vassilev *et al.* (2010, 2).

Biomass type	Volatile Matter			Fixed Carbon			Moisture Content			Ashes		
	Avg.	Min.	Max.	Avg.	Min.	Max.	Avg.	Min.	Max.	Avg.	Min.	Max.
Straws	66.7	58	73.9	15.3	12.5	17.8	10.2	7.4	16.8	7.8	4.3	18.6
Wood	62.9	30.4	79.8	15.1	6.5	24.1	19.3	4.7	62.9	2.7	0.1	8.4
Agricultural Grass	66	41.5	76.6	16.9	9.1	35.3	12	4.4	47.9	5.1	0.8	18.6
Sewage sludge	69	46.5	73.5	14.1	9.5	16.8	12.6	4.5	42	4.3	0.8	9.4
	63.7	40.9	79	8	0.5	14.5	11.6	2.5	38.1	16.7	3.2	42.3

Conclusions

Material and energy balances were conducted to propose a concept for process integration for the design of a self-sustainable fluidised bed gasifier. Using in-house developed code, theoretical calculations were done to determine the effect of moisture content in biomass used for gasification, and the plausibility of involving auto-generated char and steam in the process, seeking increased efficiency.

It was concluded that the gasification of biomass with controlled moisture content can be beneficial for the heating value of the product gas, as well as for energy savings. Moreover, assuming that the char used for catalytic reforming does not deactivate before two hours, and for the studied conditions, the amount of char produced from gasification is enough to preserve the char volume (and syngas residence ratio) in the catalytic char bed. Adequate design of a reactor should include either a char-purging system, or an additional reactor for char gasification.

The use of auto-generated steam for catalytic reforming presents an opportunity for energy savings during the steam reforming process. However, since the amount of steam generated during gasification was less than 15% volume, experiments need to be conducted to determine whether this is enough to ensure continuous tar reforming over time.

References

- Abdoulmoumine, N.; Adhikari, S.; Kulkarni, A.; Chattanathan, S. (2015). A review on biomass gasification syngas cleanup. *Applied Energy* 155, 294-307. <https://doi.org/10.1016/j.apenergy.2015.05.095>
- Abu El-Rub, Z.; Bramer, E. A.; Brem, G. (2008). Experimental comparison of biomass chars with other catalysts for tar reduction. *Fuel* 87, 2243-2252. <https://doi.org/10.1016/j.fuel.2008.01.004>
- Al-Rahbi, A. S.; Williams, P. T. (2017). Hydrogen-rich syngas production and tar removal from biomass gasification using sacrificial tyre pyrolysis char. *Applied Energy* 190, 501-509. <https://doi.org/10.1016/j.apenergy.2016.12.099>
- Basu, P. (2013). Biomass gasification, pyrolysis and torrefaction. *Biomass Gasification, Pyrolysis and Torrefaction*, 439-455. <https://doi.org/10.1016/B978-0-12-396488-5.00013-7>
- Bridgwater, A. V.; Toft, A. J.; Brammer, J. G. (2002) *A Techno-Economic Comparison of Power Production by Biomass Fast Pyrolysis with Gasification and Combustion*, 6. [https://doi.org/10.1016/S1364-0321\(01\)00010-7](https://doi.org/10.1016/S1364-0321(01)00010-7)
- Brown, R. C. (2011). Thermochemical processing of biomass: conversion into fuels, chemicals and power. <https://doi.org/10.1002/9781119990840>
- Buentello-Montoya, D.; Zhang, X.; Li, J.; Ranade, V.; Marques, S.; Geron, M. (2020). Performance of biochar as a catalyst for tar steam reforming: effect of the porous structure. *Applied Energy*, <https://doi.org/10.1016/j.apenergy.2019.114176>

- Buentello-Montoya, D.; Zhang, X. (2019). An energy and exergy analysis of biomass gasification integrated with a char-catalytic tar reforming system. *Energy & Fuels* 33, 8746-8757. <https://doi.org/10.1021/acs.energyfuels.9b01808>
- Burhenne, L.; Aicher, T. (2014). Benzene removal over a fixed bed of wood char: the effect of pyrolysis temperature and activation with CO₂ on the char reactivity. *Fuel Processing Technology* 127, 140-148. <https://doi.org/10.1016/j.fuproc.2014.05.034>
- Corella, J.; Orío, A.; Toledo, J. M. (1999). Biomass gasification with air in a fluidized bed: exhaustive tar elimination with commercial steam reforming catalysts. *Energy & Fuels* 13, 702-709. <https://doi.org/10.1021/ef980221e>
- Çengel, Y.; Cimbala, J. M. (2006). *Fluid Mechanics: Fundamentals and Applications. Fluid Mechanics: With Problems and Solutions, and an Aerodynamic Laboratory*. <https://doi.org/10.1088/1751-8113/44/8/085201>
- Dahiya, A. (2020). *Bioenergy: Biomass to Biofuels*, ISBN-13: 978-0124079090.
- Font Palma, C. (2013). Model for biomass gasification including tar formation and evolution. *Energy & Fuels* 27, 2693-2702. <https://doi.org/10.1021/ef4004297>
- Fuentes-Cano, D.; Gómez-Barea, A.; Nilsson, S.; Ollero, P. (2013). Decomposition kinetics of model tar compounds over chars with different internal structure to model hot tar removal in biomass gasification. *Chemical Engineering Journal* 228, 1223-1233. <https://doi.org/10.1016/j.cej.2013.03.130>
- Gadsbøll, R. Ø.; Clausen, L. R.; Thomsen, T. P.; Ahrenfeldt, J.; Henriksen, U. B. (2019). Flexible two-stage biomass gasifier designs for polygeneration operation. *Energy* 166, 939-950. <https://doi.org/10.1016/j.energy.2018.10.144>
- Gao, H.; Zhu, Y.; Fu, F.; Wu, H.; Du, Y.; Chen, H.; Liao, C.; Fan, H. (2014). Pyrolysis of hailer lignite in an autogenerated steam agent. *Journal of Thermal Analysis and Calorimetry* 117, 973-978. <https://doi.org/10.1007/s10973-014-3776-5>
- Gómez-Barea, A.; Ollero, P.; Leckner, B. (2013). Optimization of char and tar conversion in fluidized bed biomass gasifiers. *Fuel* 103, 42-52. <https://doi.org/10.1016/j.fuel.2011.04.042>
- Hayashi, J. I.; Takahashi, H.; Iwatsuki, M.; Essaki, K.; Tsutsumi, A.; Chiba, T. (2000). Rapid conversion of tar and char from pyrolysis of a brown coal by reactions with steam in a drop-tube reactor. *Fuel* 79, 439-447. [https://doi.org/10.1016/S0016-2361\(99\)00179-9](https://doi.org/10.1016/S0016-2361(99)00179-9)
- Henriksen, U.; Ahrenfeldt, J.; Jensen, T. K.; Gøbel, B.; Bentzen, J. D.; Hindsgaul, C.; Sørensen, L. H. (2006). The design, construction and operation of a 75 KW two-stage gasifier. *Energy* 31, 1542-1553. <https://doi.org/10.1016/j.energy.2005.05.031>
- Jess, A. (1996). Mechanisms and kinetics of thermal reactions of aromatics from pyrolysis of solid fuels. *Fuel* 75, 1441-1448. [https://doi.org/10.1016/0016-2361\(96\)00136-6](https://doi.org/10.1016/0016-2361(96)00136-6)
- Leijenhorst, E. J.; Wolters, W.; Van De Beld, B.; Prins, W. (2015). Staged biomass gasification by autothermal catalytic reforming of fast pyrolysis vapors. *Energy & Fuels* 29, 7395-7407. <https://doi.org/10.1021/acs.energyfuels.5b01912>
- Levenspiel, O. (1999). Chemical reaction engineering. *Industrial & Engineering Chemistry Research* 38, 4140-4143. <https://doi.org/10.1021/ie990488g>
- Lim, Y. Il; Lee, U. Do. (2014). Quasi-equilibrium thermodynamic model with empirical equations for air-steam biomass gasification in fluidized-beds. *Fuel Processing Technology* 128, 199-210. <https://doi.org/10.1016/j.fuproc.2014.07.017>
- Luo, S.; Guo, J.; Feng, Y. (2017). Hydrogen-rich gas production from pyrolysis of wet sludge in situ steam agent. *International Journal of Hydrogen Energy* 42, 18309-18314. <https://doi.org/10.1016/j.ijhydene.2017.04.165>
- Morf, P.; Hasler, P.; Nussbaumer, T. (2002). Mechanisms and kinetics of homogeneous

- secondary reactions of tar from continuous pyrolysis of wood chips. *Fuel* 81, 843-853. [https://doi.org/10.1016/S0016-2361\(01\)00216-2](https://doi.org/10.1016/S0016-2361(01)00216-2)
- Pereira, E. G.; Da Silva, J. N.; De Oliveira, J. L.; MacHado, C. S. (2012). Sustainable energy: a review of gasification technologies. *Renewable and Sustainable Energy Reviews* 16, 4753-4762. <https://doi.org/10.1016/j.rser.2012.04.023>
- Phuphuakrat, T.; Namioka, T.; Yoshikawa, K. (2010). Tar removal from biomass pyrolysis gas in two-step function of decomposition and adsorption. *Applied Energy* 87, 2203-2211. <https://doi.org/10.1016/j.apenergy.2009.12.002>
- Roberts, D. G.; Harris, D. J. (2007). Char gasification in mixtures of CO₂ and H₂O: Competition and inhibition. *Fuel* 86, 2672-2678. <https://doi.org/10.1016/j.fuel.2007.03.019>
- Schuster, G.; Löffler, G.; Weigl, K.; Hofbauer, H. (2001). Biomass steam gasification - an extensive parametric modeling study. *Bioresources Technology* 77, 71-79. [https://doi.org/10.1016/S0960-8524\(00\)00115-2](https://doi.org/10.1016/S0960-8524(00)00115-2)
- Sikarwar, V. S.; Zhao, M.; Clough, P.; Yao, J.; Zhong, X.; Memon, M. Z.; Shah, N.; Anthony, E. J.; Fennell, P. S. (2016). An overview of advances in biomass gasification. *Energy & Environmental Sciences* 9, 2939-2977. <https://doi.org/10.1039/c6ee00935b>
- Smith, J. M. (1950). Introduction to chemical engineering thermodynamics. *Journal of Chemical Education*. <https://doi.org/10.1021/ed027p584.3>
- Sun, Q.; Yu, S.; Wang, F.; Wang, J. (2011). Decomposition and gasification of pyrolysis volatiles from pine wood through a bed of hot char. *Fuel* 90, 1041-1048. <https://doi.org/10.1016/j.fuel.2010.12.015>
- Świerczyński, D.; Libs, S.; Courson, C.; Kiennemann, A. (2007). Steam reforming of tar from a biomass gasification process over Ni/Olivine catalyst using toluene as a model compound. *Applied Catalysis B: Environmental* 74, 211-222. <https://doi.org/10.1016/j.apcatb.2007.01.017>
- Taralas, G.; Kontominas, M. G.; Kakatsios, X. (2003). Modeling the thermal destruction of toluene (C₇H₈) as tar-related species for fuel gas cleanup. *Energy & Fuels* 17, 329-337. <https://doi.org/10.1021/ef0201533>
- Trippe, F.; Fröhling, M.; Schultmann, F.; Stahl, R.; Henrich, E. (2011). Techno-economic assessment of gasification as a process step within biomass-to-liquid (BtL) fuel and chemicals production. *Fuel Processing Technology* 2011. <https://doi.org/10.1016/j.fuproc.2011.06.026>
- Tuomi, S.; Kurkela, E.; Simell, P.; Reinikainen, M. (2015). Behaviour of tars on the filter in high temperature filtration of biomass-based gasification gas. *Fuel* 139, 220-231. <https://doi.org/10.1016/j.fuel.2014.08.051>
- Vassilev, S. V.; Baxter, D.; Andersen, L. K.; Vassileva, C. G. (2010). An overview of the chemical composition of biomass. *Fuel* 89, 913-933. <https://doi.org/10.1016/j.fuel.2009.10.022>
- Wang, Y.; Yoshikawa, K.; Namioka, T.; Hashimoto, Y. P. (2007). Performance optimization of two-staged gasification system for woody biomass. *Fuel Processing Technology* 88, 243-250. <https://doi.org/10.1016/j.fuproc.2006.10.002>
- Woolcock, P. J.; Brown, R. C. (2013). A Review of cleaning technologies for biomass-derived syngas. *Biomass and Bioenergy* 52, 54-84. <https://doi.org/10.1016/j.biombioe.2013.02.036>
- Xiong, S.; Zhuo, J.; Zhang, B.; Yao, Q. (2013). Effect of moisture content on the characterization of products from the pyrolysis of sewage sludge. *Journal of Analytical and Applied Pyrolysis* 104, 632-639. <https://doi.org/10.1016/j.jaap.2013.05.003>
- Zeng, X.; Dong, Y.; Wang, F.; Xu, P.; Shao, R.; Dong, P.; Xu, G.; Dong, L. (2016). Fluidized bed two-stage gasification process for clean fuel gas production from herb residue: fundamentals and demonstration. *Energy & Fuels* 30, 7277-7283. <https://doi.org/10.1021/acs.energyfuels.6b00765>

Zhang, X. (2016). Essential scientific mapping of the value chain of thermochemically converted second-generation bio-fuels. *Green Chemistry*

18, 5086-5117. <https://doi.org/10.1039/C6GC02335E>