Hybridization of biomass steam gasification in dual fluidized bed reactor with concentrated solar energy

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Abstract

Production of syngas from biomass gasification using concentrated solar energy is an attractive technology to produce storable renewable energy and CO_2 reduction. Although many solar gasification reactors have been conceptually developed during the last decades, none has reached commercial status yet. Several challenges have hindered the deployment of the technology due to difficulties derived from: high temperature and/or large reactor volume required for complete fuel conversion; producing a steady syngas output independent on solar radiation variation; achieving effective rate of heat supply to the gasifier when scaling-up. In this paper, a new approach to conduct allothermal gasification of biomass with steam using concentrating solar energy with the potential to overcome the aforementioned technical challenges is presented. It is based on the use of solid particles as thermal energy carrier and storage media. The great advantage of this configuration is that the solar receiver and the reactor are uncoupled, while thermal integration is highly efficient since carrier particles are directly used in the reactor. This paper reviews the state of the art of the most important aspects to develop the technology and presents a model to preliminary analyze the performance of the reactor under various configurations.

1. Introduction

The use of solar energy as external heat source for steam reforming of fuels has been recognized as highly attractive method for increasing the share of renewable energy and reduction of CO_2 emissions. The generation of energy vectors carrying the energy from the sun and the biomass, is an encouraging concept towards full renewable energy production and energy storage. Solar steam gasification of biomass is one of the most attractive technologies considered for the achievement of such objectives.

Steam gasification of biomass proposed until now are either autothermal (the heat is supplied by partial burning of the fuel) or just laboratory studies where the heat is supplied by electrical heaters. The external heat source would be ideally carried out by transferring all the energy available in the fuel into the produced syngas (instead of burning part of the fuel to provide the heat), hence raising the yield significantly and improving the overall energy efficiency of the process. However, it is vet to be determined how to supply the solar heat to industrial scale reactors. Various conceptual strategies to hybridize solar energy into steam gasification of biomass have been recently proposed by [1], examining how to the authors continuous implement allothermal gasification using solid particles to carry and store the solar energy.

The objective of this paper is to examine the progress on the solar gasification design based on the new concept. A model for new gasification system is developed previous based on a model for conventional fluidized bed gasifiers (FBG) [2] and first calculations are presented to analyze optimal operational conditions. The results are useful to identify the main challenges, shedding light on the next steps for the design of the first generation of reactors based on this concept.

2. Background

Prior to introducing the new concept, the state of the art of solar and non-solar steam gasification and concentrating solar thermal energy for high temperature applications based on solid particle receivers is given in order to justify the new prototype.

2.1. Steam gasification of biomass

The steam gasification of biomass (dry and ash free, termed as "bio" with general formula $CH_{1.44}O_{0.66}$) to yield syngas (H₂/CO) or (H₂/CO₂) can be represented by [3]:

$$\begin{array}{l} {\rm CH}_{1.44}{\rm O}_{0.66} {+}0.34\,{\rm H}_2{\rm O} {\rightarrow} 1.06\,\,{\rm H}_2 {+}\,{\rm CO} \quad ({\rm R1}) \\ {\rm CH}_{1.44}{\rm O}_{0.66} {+}1.34\,{\rm H}_2{\rm O} {\rightarrow} 2.06\,\,{\rm H}_2 {+}\,{\rm CO}_2 \ ({\rm R2}) \end{array}$$

The standard heat of reaction at 298 K of R1 and R2 are, respectively, 102.5 and 61.3 kJ/mol_{bio} or 4.27 and 2.55 MJ/kg_{bio}. Therefore, gasification is an endothermal process, requiring significant input to drive the reactions. In addition, because of equilibrium and kinetic reasons, the process must be conducted at high temperature, in the range of 800-1100 °C depending on the fuel and reactor type.

Gasification technologies include allothermal steam gasification, using heat from an external source to drive the process, and autothermal gasification, in which part of the fuel is burnt to generate the necessary heat to maintain the process, using pure oxygen or air. Since in autothermal gasification part of the fuel is consumed to provide thermal heat, the efficiency is lower than in allothermal gasification.

Despite a great number of allothermal steam gasifiers has been developed at laboratory or bench scale (some of the reviewed in [3,4] none has been scale up or commercialized due to practical problem derived from steady production of syngas in spite of solar radiation variation as well as implementation of heat transfer at large scale.

Therefore, steam (non-solar) gasification has been implemented at scale by the use of indirect air-steam dual fluidized bed (DFBG) technology [4], i.e. bv autothermal gasification using air. In this arrangement, the biomass is devolatilized in a bubbling FBG where volatiles are released, and char is partially gasified (\approx 5-30% depending on the fuel) with large excess of steam. The process is thermally driven by the circulating hot material from the circulating FB combustor (where the char transported from gasifier and an additional fuel, if necessary, are burnt to heat the sand material). The bubbling FBG

unit is allothermal since the heat from sand comes from the combustor unit (external to the gasifier), although the "gasification system" as a whole (gasifier+combustor) is autothermal. As a result, only about 70% of the energy (and 2/3 of the carbon) from the fuel is stored as chemical energy in the syngas. This is the usual way to operate the existing DFBG as developed by TUV, ECN or some universities/research centers and commercialized by companies like Repotec or Dahlman, although sometimes these DFBG operated burning also natural gas to elevate the combustion temperature.

In Fig. 1, the possibilities of the classical air-steam DFBG are extended bv considering that the system can be balanced thermally by some external heat or additional fuel. For obvious reasons, the most attractive case is when all external energy supplied to the gasifier is renewable (i.e. solar) and the fuel is a biomass. In this case the solar energy is transferred to the bio-syngas and all carbon from the biomass is converted to fuel volatiles (CO and hydrocarbons). In turn, it produces a syngas with roughly 115% of the energy and 100% of the carbon of the fuel. Despite the great advantage of this operational mode, sustaining the gasifier with external heat is of most complexity and has not been developed. The approach proposed in the present paper is to convert the existing conventional design of DFBG to admit external solar heat, maximizing the solar share in the syngas.



Fig.1: Steam gasification in a dual fluidized bed (DFB) using external heat or fuel

2.2. Solar gasifiers

Solar gasifiers can be classified according to the gas-solid contact (packed bed, fluidized bed, and entrained flow gasifiers) or by the way in which solar radiation contacts the reactants (directly-irradiated, where the solid carbonaceous reactants are exposed to radiation. directly and indirectly-irradiated, where the radiation strokes an intermediate material -opaque wall or energy carrier-). A variety of combinations of gas-solid and solar radiation-reactants contact reactors have been proposed, and some prototypes have been tested at laboratory scale [5,6].

Direct irradiation offers superior heat characteristics and transfer energy efficiency, but the reactor must have a transparent window, which can be fouled by operation. Indirect irradiation, in which an intermediate medium is heated by solar radiation and transported to the gasifier, has less favorable heat transfer but avoids the difficulties with direct-irradiation design [7]. Packed-bed gasifiers are simpler and robust, can accommodate a wide range of feedstock sizes, making them cost effective, but they suffer from mass and heat transfer limitations, ash build-up, and energy losses. Entrainedflow gasifiers exhibit more efficient increasing transport, the syngas throughput significantly, but impose strict requirements on the feedstock size. Both directly- and indirectly- irradiated have been developed for all gas-solid contacts [5,6].

Fluidized-bed reactors achieve high mass and heat transfer rates, overcoming the transport limitations of packed-bed and the particle size needs of entrained-flow reactors, being considered the solar gasifier with highest scaling-up potential although some issues remain to be resolved. An earlier design based on a conventional bubbling FB was modified into an internally-circulating FB to improve reactor performance, achieving more uniform bed temperature, although the performance was questionable and scaling-up remains unsolved [6]. Indirectly-heated FBG have been proposed to overcome these difficulties, including three approaches to indirectly supply the concentrated solar radiation (CSR) to the reactor: (i) irradiating the reactor external side walls [8]; (ii) using a two-cavity reactor [9]; and (iii) using solid particles as heat transfer carrier [10]. The only reported theoretical investigation based on solar gasifier heated by circulating solids was done in [10,11]. In [10] a hypothetical hybridized DFB gasifier to study the production of Fischer-Tropsch liquid from coal was tentatively considered. In [11] a lift pipe as part of a fluidized-bed was proposed where the bed material and char are heated by heat pipes which are externally irradiated. The gasification proposed in [10] is autothermal since the char and some auxiliary fuel are burnt, decreasing significantly the solar share in the syngas and the syngas yield per unit of fuel input. In addition, no experimental support of this concept has been published.

To sum up, none of aforementioned directly-irradiated gasifiers can be implemented at scale and, in the best cases, only lab-scale devices have been tested to understand the process limits of gasification subjected to solar radiation. The only existing FB solar gasifier lab-scale reaching the is directlyirradiated, but the performance is poor and cannot be applied for large scale applications. The use of solids particles as energy carrier for gasification has theoretically proposed in two earlier works, but has not been experimentally tested.

2.3. Solid particle solar receivers

Solid particles were proposed as heat transfer and thermal storage medium in the early 1980s because of its ability to withstand high temperatures and the straightforward integration of solar energy collection and TES, but the technology plummeted very fast. Only in recent years, driven by the need of achieving higher temperatures and efficient together with cheap TES, solid particle technology has been subject of new research interest [12].

The heat transfer mechanism at the receiver can be either direct or indirect. In the direct type, the solid particles are directly irradiated by CSR, whereas in the indirect type solid particles 'flow' inside tubes. Early direct absorption concepts considered a curtain of free-falling particles. Obstructed or impeded flow receivers, rotary kiln and fluidized bed receivers have been proposed to increase residence time of the particles. Regarding indirect heat transfer concepts, flow-intube receivers. with and without fluidization, have been tested at small scales. Some technical challenges that must be solved for commercial viability of solid particle technologies [12].

Integration of solar energy conveying by solids particles coming from the hot storage (storing hot particles from the central receiver) is usually conducted by heat exchanger to produce steam or to heat an additional HTF [13]. New solid particle receivers are under development toward more efficient thermodynamic cycles based on gas turbines (Brayton cycle). No thermochemical applications have been considered so far under this operational mode, except the two theoretical studies mentioned [10,11].

2.4. Main conclusions from literature

- Allothermal gasification with external heat from concentrated solar radiation is the most interesting choice for steam gasification of biomass since the syngas produced contains all energy from the biomass and a significant solar share. In addition, it contains most of the carbon of the biomass as combustible species. In spite of this, allothermal steam gasification has been tested only in the laboratory with heat from an external electrical oven.
- The only practical development on steam gasification (no-solar) is the air-steam indirect DFBG. In this technology the heat to the gasifier is achieved by burning part of the fuel (char) and providing this gasifier with external heat has not been considered. As a result, there is not a solar version of the technology.
- Most of solar gasifiers developed up to date are directly-irradiated. They are more efficient that indirectly irradiated gasifiers but scaling-up remains a challenge. As a result, only laboratory devices have been tested. Direct irradiation to the reactor should be avoided in order to overcome all problems hindering application at large scale.
- New technology of solids particle receivers (without reaction) reaching temperatures between 700-900°C has been developed in the last 5 years. Moreover. research under development promises good future perspectives to come up with scalable prototypes reaching 1000°C. The directly-irradiated solar particle, tower-mounted, falling particle cavity receiver is the best choice developed to date and can achieve the thermal requirements of biomass the gasification (800-950°C).

- The solid receiver and the gasifier can be uncoupled using solid particles as energy carrier and storage material. However, excessive storage volume and complications in conveying great amount of solid particles.

The conceptual integration of the solar gasification system and particle receiver is presented in Fig.2. The solids particles act as thermal energy carrier, circulating cyclically between the solar receiver and the gasifier. Two tanks are used to store the particles heated by the receivers and the particle cooled in the gasifier allowing for temporary thermal storage of solar energy. The use of solid particles as energy carrier is attractive since are an excellent thermal energy storage medium, operating at high temperature and low cost.



Fig.2: Integration of biomass gasifier in a solar loop with a solid particle receiver

3. Modelling

3.1. Model approach

The stoichiometry of reactions R1 and R2 establishes the amount of reactants involved (steam per unit of biomass or SBR) and products (H₂ and CO/CO₂) generated provided the reactions are complete and stoichiometric. This is not the case in practice because of thermodynamic and kinetic limitations, yielding a more complex product distribution [2].

Three approaches can be applied to model steam gasification in an FBG: (i) The assumption of equilibrium (EM); (ii) application of kinetics models (KM), taking into account chemical and fluiddynamics rate considerations; (iii) a combined approach, sometimes called pseudo-equilibrium (PEM). Equilibrium model (EM) is the most universal way to close the calculations but fails in predicting real gas composition and fuel utilization (char conversion). Kinetic model (KM) gives better representation of the process for a specified system (geometry, type of biomass, etc.) but a great deal of inputs is required and the conclusions system-dependent. are Pseudo-equilibrium model (PEM) is based on equilibrium relations together with semiempirical inputs to take into account kinetic- and flow- rate limitations. It is a reasonable compromise between EM and KM using some comprehensive models supported by empirical closures.



Fig.3: Steam gasification in a solar-heated dual fluidized bed gasifier (sh-DFB)

The model of the steam gasification of a DFBG using heated solids particles (sh-DFBG) (Fig.3) considers the steam gasification in a FBG and the char burning in the combustor. In both units a circulation of solids (including those coming from the solar loop) flowing in and out must be considered as discussed

below. In this system it is convenient to define two types of solids circulations: internal solids circulation (between reactors) and external circulation (that from the solar loop).

3.2. Model of a solar-heated DFBG

The steam gasification process taking
place in the gasifier is represented by
$$CH_{1.44}O_{0.66} + \lambda_{H_2O} H_2O \rightarrow$$

 $n_{H_2}H_2 + n_{CO}CO + n_{CO_2}CO_2 +$ (R3)
 $n_{CH_4}CH_4 + n_{C_2H_4}C_2H_4 +$
 $n_{C_{(S)}}C_{(s)} + n_{H_2O}H_2O + n_{C_{10}H_8}C_{10}H_8$
where besides CO and H₂ other
components are present in the product gas,
such as hydrocarbons (mainly CH₄, and
other light hydrocarbons, here lumped into
 C_2H_4 in R3) and tars (lumped into
naphthalene $C_{10}H_8$ as tar model in R3) as
well as solid carbon (char, i.e. C_(s) in R3).
In addition, there is always unconverted
steam even when feeding it
stoichiometrically.

In a stand-alone allothermal steam gasifier it is difficult to fully convert the solid carbon with steam in the gasifier. Operation at high temperature and residence time of the char is required, or the use of catalyst. In a DFBG, in contrast, only a limited fraction of the char generated after fuel devolatilization is converted with steam, being the rest directed to the char combustor, where it is burned with air, generating the heat necessary to balance the system energetically.

The extent of char conversion in any gasifier depends on the carbon-steam gasification rate of the char particles (CO_2 -carbon rate is much slower) and the residence time of the char particles in the reactor. On the one hand, the rate of reaction depends on the temperature, the

species concentration (mainly steam, but hydrogen can be also important as it inhibits the carbon-steam reaction rate), the intrinsic reactivity of the char (fuel type and form of char generation) and the quality/extent of gas-solid contact. On the other hand, the residence time of the char particles depends on the rate of solids removal, that can be (i) intentionally made by gas-solid entrainment and elutriation and/or by removing the solids in the case of DFBG to carry the solids to the combustor, or (ii) unintentionally as carbon losses by entrainment of elutriated particles from the bed in a stand-alone gasifier. Theoretically, in a gasifier with any kind of char removal, the residence time of char is infinite and the char conversion is complete, but also the volume of the reactor is very high (infinite for an infinite time of complete carbon conversion in a single particle).

The model of char conversion developed in [2] for a FB is applied in the present reactor model. The char conveys to combustion is burn with air according to

$$\begin{array}{l} n_{\mathrm{C(s)}} \mathrm{C(s)} + \lambda_{\mathrm{O_2}} \mathrm{O_2} + 3.76\lambda_{\mathrm{H_2O}} \ \mathrm{N_2} \rightarrow \\ n_{\mathrm{CO_2,s}} \mathrm{CO_2} + 3.76\lambda_{\mathrm{O_2}} \ \mathrm{N_2} \end{array}$$
(R4)

3.3. Process configurations

As said, in a standard (no-solar) DFBG there is only internal solids circulation between the units. In contrast, in sh-DFBG besides internal circulation, there is also external solids circulation (through the solar loop). Furthermore, in a sh-DFBG there are different possibilities for introducing/extracting the solids in the system as show in Fig.4, and the internal circulation from the gasifier to the combustor can be different to that from the combustor to the gasifier. The biomass spatial time defined as mass of inventory in the gasifier divided by the feed rate of biomass is always higher in the sh-DFBG compared to conventional DFBG because higher throughput of inert particles (those solids different from fuel and char) passing through the gasifier.



Fig.4: Options for feeding/extracting the solids in an sh-DFBG

3.4. Performance indicators

To quantify the contribution of external solar energy to the system, we define the following parameters:

- Specific External Heat (SEH) defined as the solar heat supplied to the system Q_{SL} per unit of biomass input (dry and free of ash), (MJ/k_{daf}).
- Fraction of external heat supplied to the system (FEH) defined as the ratio between Q_{SL} and the heat required to thermally-sustain the gasifier Q_g . FEH= Q_{SL}/Q_g .
- *Solar Share* (SS), defined as ratio between Q_{SL} and the lower heating value of the syngas, expressed in percentage, SS=(Q_{SL}/LHV_{syngas})·100

4. Results and discussion

Fig. 5 presents the performance of a standard (non-solar) DFBG at different gasification temperatures for fixed combustion temperature, steam equivalence ratio ER_{H2O} (the ratio between the fed and stoichiometric steam), and inlet steam temperature. It is shown that the solids circulation increases as the

gasifier operates at higher temperature. Consequently, lower char residence time is required and lower char conversion is attained in the gasifier (Fig. 5(a)). The yields of syngas and H₂ (Fig. 5(b)) decrease with gasification temperature as a result of the lower char conversion in the gasifier.

Fig. 6 shows the performance of a sh-DFBG operating at fixed gasification temperature with solids addition/removal to/from the gasifier (Confl, see Fig. 4), as a function of the fraction of external heat supplied to the system, FEH. The internal solids circulation decreases significantly with FEH, whereas the biomass spatial time τ_{bio} (larger mass inventory or reactor size for a given biomass flowrate) (Fig.6(a)). Logically, increases no circulation is necessary when all the heat required to the gasifier is supplied externally (FEH=1). Significant char conversion is reached for reasonable long residence time (80% of char conversion is attained in the gasifier with residence time of 28 min) but higher char conversion requires excessive long residence time and thus reactor volumes (see τ_{bio} in Fig.6(a)). produced is improved The syngas considerably (Figure 6(c)) as a result of higher steam-char conversion. Fig. 6(d) shows that 15% of solar share can be attained for FEH=1, meaning that 115% of the energy of biomass is transferred to the syngas as compared to 70% in a conventional DFBG, thus a relative increase of 65% ((115-70)/70.100).



Fig.5: Performance of a standard (no-solar) DFBG as a function of gasification temperature (T_{gas}) for: T_{comb}=905 °C, ER_{H2O}=2, biomass type CH_{1.4}O_{0.7} and inlet steam temperature 750°C



Fig.6: Performance of a sh-DFBG with various levels of external heat ratio (FEH) for Configuration 1 at T_{gas} =850°C and T_{SL} =950°C (the rest of operating conditions as in Fig. 5)

Fig. 7 shows the external solids circulation (that circulating through the solar loop) as a function of the specific external heat SEH for the four configurations presented in Fig.4. Configurations with the solids removal in the same unit (Confs 2 and 4 and Confs 1 and 3) present the same value of external solids circulation, as this results from the driving force of temperature between the hot particles and removal point. Therefore, the external solids circulation is higher in Confs 2 and 4, since the driving force is lower.





Fig.8 compares the internal circulation for the four configurations in Fig.4 Confl requires the lowest internal solids circulation for equal specific external heat, whereas Conf4 demands the highest one. Confs 2 and 3 present different internal circulations depending on the direction of the solids flow. Moreover, Confs 1 and 3 present the highest external solar heat absorption capacity (the highest SEH), reaching a value of 3 MJ/kg_{bio}, corresponding to a solar share of 15%, as shown in Fig. 6(d) for FEH=1. In Confl the internal solids circulation in the two directions is the same and decreases with SEH, while in Conf3 the solids flow to the gasifier is higher than that to the combustor, and the difference of the two solids flows increases with FEH. The internal solids circulation in Confl becomes zero when the system reaches the maximum solar share, i.e. when it becomes completely allothermal. In this case, all the external solids pass through the gasifier only, full char conversion is attained in it, and the combustor is out of service.



Fig.8: Internal solids circulation in a sh-DFBG as a function of specific external heat supplied to the system (SEH) for different configurations (1-4) (operating conditions are the same as in Fig. 6 and 7). Nomenclature: "to gas" is from combustor to gasifier; "to comb" is from gasifier to combustor

An operational point is identified in Conf2 where the system is saturated to absorb more external solar heat, even if more external hot solids were introduced in the gasifier. At that point (SEH \approx 1 MJ/kg_{daf}) part of the char is burnt and char conversion in the gasifier is limited to 0.46 reaching a maximum solar share of about 6%. The maximum SEH attainable in Conf4 is around 2.3 MJ/kg_{daf}, lower than the maximum SEH in Confs 1 and 3 (3 MJ/kg_{daf}). This results from the difference between the driving forces of temperatures particles-combustor, (hot i.e. 950-905=45°C in this simulation) and that (gasifier-combustor, i.e. 905-850=55°C in simulation). this Since the latter temperature driving force is higher than the former, some char is burnt in the combustor to heat up the solids from the gasifier to combustor and the char conversion is lower than unity in the gasifier, resulting in SHE lower than the maximum. For the same driving forces, for instance by taking combustor temperature at 900°C (the two driving force would be 50°C), full char conversion is attained in the gasifier and SEH=3 MJ/kg_{daf}.

From the above analysis (gasification perspective) Confl is the most attractive option. However, additional aspects from solar receiver side have to take into account. Solids removal from the gasifier (Confs1 and 3) will result in reacting particles in the solar loop, which in principle poses new problems (the solid particle receivers currently under developments are mainly based on "open" designs [12]). On the other hand, removal of solids from the combustor (Confs 2 and 4) will make more difficult the solar receiver from the thermal point of view (higher temperature of the hot solids to the receiver) and will increase the circulation of solids between reactors, although it has the advantage of allowing the use of open solid particle receivers. A way to relax the disadvantage of the high thermal level is the cooling of solids from the combustor in an intermediate exchanger before sending them back to the warm solids tank. The heat exchange could generate superheated steam to feed the gasifier, for instance; however, there are other ways to superheat

the steam like exchanging with the produced syngas or flue gas and the options should be considered. The use of pipes working between heat this intermediate exchanger and the gasifier could also benefit the process as should be into account additional taken as alternative. Further research is necessary.

5. Conclusions and future work

A new conceptual integration of solar gasification and particle receiver is presented and analyzed in this paper. The solids particles act as thermal energy carrier, circulating between the solar receiver and the gasifier with intermediate thermal energy storage. The advantage of this configuration is that the solar receiver and the reactor are uncoupled, while thermal integration is highly efficient since carrier particles are directly used in the reactor. The new system can be accomplished with the state-of-the-art technology by considering some new aspects for the particular reactor conditions. Moreover, temperatures in the receiver are realizable using latest developments of solar concentrators, although some specific aspects need to be developed for an efficient matching between the biomass and solar components.

The solids circulation and quality of syngas for different arrangements have analyzed this been in work by development of a model of a solar-heated DFBG. Addition of solids to the gasifier seems to be the most attractive option but limits the operation to close solids particle receiver or enforced carrying out solids separation before sending them to the receiver. However, additional aspects have to take into account and research is underway. Optimization of the system for the different configurations is under study, as well as how to adapt the new gasifier and combustor as more external heat is

added. Criteria for optimization includes taking into account technical viability, thermal efficiency, solar share into syngas, and operational flexibility. The latter criterion includes considering stand-alone operation without solar energy, transient from solar to non-solar mode, hybrid operation and the size of the thermal storage.

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