Experimental and CFD investigation of inert bed materials effects in a high-temperature conical cavity-type reactor for continuous solar-driven steam gasification of biomass

Houssame Boujjat^{1,2}, Sylvain Rodat³, Srirat Chuayboon³, Stéphane Abanades^{3*}

¹CEA-LITEN Solar and Thermodynamic Systems Laboratory (L2ST), F-38054 Grenoble, France

²Univ. Grenoble Alpes, F-38000 Grenoble, France

³Processes, Materials and Solar Energy Laboratory, PROMES-CNRS, 7 Rue du Four Solaire, 66120 Font-Romeu, France

*Corresponding author: stephane.abanades@promes.cnrs.fr

Abstract: Solar biomass gasification was performed in a high-temperature conical bed reactor for thermochemical syngas production, which represents an efficient route to promote biomass valorization while storing intermittent solar energy into carbon-neutral solar fuels. The developed solar reactor is based on a conical cavity-type receiver enabling continuous biomass feedstock injection under real concentrated solar irradiation. In this study, the use of inert bed materials is considered to directly absorb a portion of the entering solar power and to transmit thermal energy to the gas and reactive particulate phase by radiation, convection and particle-to-particle interactions, with the aim of homogenizing the reactor temperature. A fluid dynamics study of the gas/particle flow was first performed to provide insights into the reactor hydrodynamics in the case of empty cavity and cavity containing inert particles in the forms of packed-bed and spouted-bed. The beneficial effect of the inert particles bed was emphasized. The spouted bed particles play a role in retaining the reacting biomass particles longer inside the reaction zone and promote phase mixing, as also confirmed by cold tests on a replicated transparent mockup cavity. Different inert particle bed materials were then considered and experimentally tested to unravel their impact on the biomass conversion, syngas composition and yield, and gasification performance in the solar-heated reactor operated at 1200°C and 1300°C.

Keywords: Solar energy, gasification, biomass, syngas, solar fuel, spouted-bed reactor.

1. Introduction

Over the last few decades, the interest in biomass gasification has increased due to the growing demand for reducing global greenhouse gas emissions responsible for global warming (Safarian et al., 2019). Major advantages of biomass gasification over combustion include its potential to produce different forms of energy, fuels and chemicals. In conventional industrial autothermal gasification reactors, at least 30% of the initial feedstock is burnt to supply the required heat (Higman, 2013). This reduces the complete use of the biomass for fuel production and leads to the contamination of syngas by combustion by products. The use of concentrated solar power (CSP) to drive the endothermal gasification reactions results in saving biomass resources, thus producing more syngas with a higher quality while avoiding combustion, thereby eliminating the need for downstream gas processing and pollutants abatement. Concentrated high-flux solar energy systems (e.g., solar beam-down (Noone et al., 2012) and solar towers (Li et al., 2015)) can be used to achieve high gasification temperatures ($>1200^{\circ}\text{C}$), which favours the reactions kinetics and tars thermal cracking, while being suitable to convert a wide variety of biomass feedstock types.

Solar thermochemical reactors generally depend on the method of heating the solid reactants. Directly irradiated solar gasifiers generally make use of a transparent window that allows the concentrated sunlight to enter directly the reaction chamber. In such configuration, efficient solar absorption is achieved and heat losses are lowered, which enables to reach and maintain high temperatures ($1000\text{--}1500^{\circ}\text{C}$) (Puig-Arnavat et al., 2013). However, the transparent window mechanical resistance and cleanness are restraining factors due to inherent limitations regarding pressure and particles/smoke soiling/deposition (Loutzenhiser et al., 2017). Alternatively, indirectly irradiated solar reactors use an opaque heat transfer wall to capture the highly concentrated solar flux and then transfer it to the reaction zone, thus avoiding the need for a transparent window. The type of material selected for the emissive wall should be constrained by its resistance to thermal shocks and maximum operating temperature, while it should offer chemical inertness, radiative absorbance, high thermal conductivity, and suitability for transient operation. The first solar gasifier (Gregg et al., 1980) was a directly irradiated fixed bed reactor and was successful to convert different carbonaceous materials into syngas with more than 40% of the incoming sunlight effectively stored as fuel value in the product gas. Taylor et al. (1983) studied a directly irradiated fixed bed reactor for charcoal, wood and paper gasification. This reactor was irradiated from the top and the charge was pushed upwards the focal point using a piston as the gasification progressed. The obtained performance was compared to that of a directly irradiated fluidized bed reactor for CO_2 charcoal gasification. Compared to its fixed bed equivalent, which has an energy efficiency of 40%, the fluidized bed reactor reached only 10% efficiency at 950°C because of the more pronounced radiation losses on the upper part of the reactor and additional sensible losses in the exit gas. Piatkowski et al. (2009) studied another fixed bed reactor with batch mode operation. This reactor was indirectly irradiated using a solar simulator and was used to convert a wide variety of carbonaceous waste feedstocks such as

scrap tire powder, industrial sludge and sewage sludge. The main drawback of this reactor was its very low conductive heat transfer rate that entailed significant temperature gradients and impaired the conversion throughout the bed. Kodama et al. (2002) gasified bituminous coal with CO₂ in a small-scale fluidized bed reactor directly irradiated by a Xe lamp from the side. The fluidized coal particles were in direct contact with the quartz window, which was detrimental to its cleanliness and mechanical integrity. Therefore, in another study, the concept was improved by developing a new solar fluidized bed reactor irradiated from the top to insure a gap between the window area and the reacting particles (Kodama et al., 2010). This fluidized bed reactor was further improved (Gokon et al., 2012) by adding a draft tube in the centre of the reactor, which allowed homogenizing the temperature throughout the bed. Entrained and vortex flow reactors were also solarized, and tested under both direct and indirect heating modes. Z'Graggen et al. (2006) studied directly irradiated petroleum coke steam-gasification. The vortex flow configuration allowed achieving up to 87% of petcoke conversion in a single pass of 1s residence time. In the same vein, Müller et al. (2017) gasified charcoal-water slurry in a windowless indirectly irradiated SiC cavity reactor at elevated pressure ranging from 1 to 6 bar. A reaction extent of 94% was obtained in less than 5s with an upgraded calorific value of the feedstock of 35%. Other reactor designs were investigated such as two-zone drop tube/packed bed reactors (Bellouard et al., 2017a; Kruesi et al., 2013) where a porous material was placed in the hot region of the reactor to increase the particles residence time in the reaction zone, and molten salt reactors (Hathaway et al., 2014) in which molten salt was used as both heat transfer medium and catalyst for the reaction.

The present study was undertaken following the results of a previous work (Boujjat et al., 2019a,b) in which a novel 1.5 kW_{solar} dilute solar spouted bed reactor was simulated and experimentally studied for the first time. In the preceding work, the reactor was initially empty and the continuously injected biomass particles were the only spouted particles inside the cavity. Such technology was suitable to continuously convert the biomass feedstock with a high carbon conversion efficiency (~80%), and the efficient storage of solar energy in the form of a readily transportable fuel was confirmed. With the support of 3D multiphysics modelling, improvement strategies were proposed to enhance the phase mixing, to limit the finest char particles entrainment and to homogenize the reactor temperature. The use of inert particles (set at the bottom of the conical cavity in the form of spouted or packed-bed) appeared to be judicious to achieve higher reaction extents and heat transfer rates. Therefore, in the present work, different inert bed materials were tested for the first time to study their impact on the allothermal (solar-only) continuous gasification of wood particles. Prior to the experimental study, the gas/particles flow dynamics at real processing condition was investigated using Computational Fluid Dynamics (CFD) modelling. Two thermal CFD models were developed on ANSYS-Fluent v19.1 to simulate the flow through the inert packed and spouted bed particles in interaction with radiation and gas. These simulations were completed by cold tests on a replicated transparent PMMA (Polymethyl methacrylate) cavity mockup to validate the CFD predictions (at ambient temperature) and confirm the effective mixing

of the wood and bed particles. Thereafter, high temperature solar experiments were carried out at the CNRS-PROMES solar furnace under real direct concentrated solar flux. Results in terms of syngas yield, composition, biomass conversion and reactor performance metrics were discussed in light of the numerical simulations.

2. Experimental set up

2.1 Solar reactor

The solar reactor was settled at the focus of a vertical-axis parabolic facedown concentrator (Supporting Information, Figure SI1). The reactor was based on the principle of conical spouted beds (Figure 1). Being conical at the bottom and cylindrical in the upper free board region, a high velocity inlet fluid flow expands the bed, and then spouts the central particles from the bottom to the upper fountain (Bellouard et al., 2017b, 2018). A systematic circulation pattern is achieved in the upward direction through the spout and slowly downward in the annulus region. Spouted bed reactors have been developed for various applications including drying (Markowski and Kaminski, 1983), pyrolysis (Makibar et al., 2012) and non-solar gasification (Lopez et al., 2017). Previous works on gasification have revealed the interests of spouted beds over conventional fluidization for converting cohesive and sticky particles of various sizes, shapes, densities, and textures (Erkiaga et al., 2013; Mathur and Gishler, 1955; Olazar et al., 1992, Chuayboon et al., 2018a,b).

Figure 1. Scheme of the solarized conical dilute spouted bed reactor

Table 1 summarizes the main reactor materials and their physical properties. The cavity receiver was consisting of a vertical cylinder (78 mm inside diameter) with a 60° angle conical bottom (total cavity

height: 115 mm) and was made of a high-temperature resistant metallic alloy (FeCrAl). The bottom inlet of the cavity was used for injecting the spouting gas via an alumina tube of 2 mm inside diameter from which the oxidizing agent (water steam) was fed. A layer (30 mm-thick) of porous ceramic fiber (aluminosilicate) was used to insulate the whole cavity. An alumina cap (20 mm-diameter aperture) was placed above the metallic cavity to maximize incoming absorbed solar radiation while reducing heat losses. A layer of zirconia felt (2 mm thickness) and a protective graphite plate were placed above the alumina cap to protect it from the direct high-flux solar radiation and minimize radiation losses. A stainless steel screw feeding system coupled with a hopper was used to transport the feedstock directly into the cavity. The whole feeding system was hermetically fixed to the water-cooled reactor shell.

Table 1. Reactor materials and properties (at 1400°C otherwise specified)

Component	Density ρ (kg.m ⁻³)	Heat capacity c_p (J.kg ⁻¹ .°C ⁻¹)	Thermal conductivity λ (W.m ⁻¹ .°C ⁻¹)
Alumina cap	4000	795	7
FerCrAl alloy	7100	800	35
Insulation	400	800	0.22
Zirconia layer	5700	500 (25°C)	2.2 (20°C)
Graphite layer	1400	710 (25°)	25 (25°C)

2.2 Methods

The gasified beech wood particles had an average diameter of 1 mm (Figure 2). Their characteristics are summarized in Table 2. They were obtained from XP CEN/TS 15104, XP CEN/TS 15105, XP CEN/TS 15107, XP CEN/TS 14775 standards. Oxygen content was deduced by difference. The LHV (dry basis) was measured with a PARR 6200 bomb calorimeter knowing humidity from NF EN 14774 standard. Granulometry was obtained using a FT4 granulometer.

Table 2. Biomass composition and properties

C (wt.%)	H (wt.%)	O (wt.%)	N (wt.%)	S (wt.%)	Ash (wt.%)	Moisture (wt.%)	Density (kg.m ⁻³)	Specific heat (J.kg ⁻¹ .°C ⁻¹)	LHV (MJ.kg ⁻¹)
48.3	6.7	44.4	0.1	<0.1%	0.46	8.9	650	1500	16.93

The feedstock was stored in a 1.15L hopper and was continuously injected by the screw feeder. The wood particles were transported into the hot cavity receiver and fell by gravity until complete injection of the loaded biomass particles.

A flow of Ar (0.5 NL/min) was continuously injected via the hopper through the feeder, so that the hot gases from the cavity could not interact with the reactive biomass feedstock. Moreover, to avoid window fouling and overheating, a protective Ar flow (2 NL/min) was constantly flowed below the window for sweeping and ensuring that all the product gases are directed to the reactor outlet. The jet gases (a mixture of Ar carrier gas and steam) were injected from the alumina tube at the cavity bottom. The composition (0.2 NL/min of Ar and 200 mg/min of steam) was adjusted to provide a slightly over-stoichiometric Steam-to-Biomass (S/B) ratio.

The produced gases flowed through a bubbler and two cartridge filters to eliminate excess steam and entrained char particles. An on-line gas analyzer (GEIT 3100) was used to continuously monitor the syngas molar composition. The H₂ mole fraction was measured by a thermal conductivity detector (TCD) while the mole fractions of CO, CO₂, and CH₄ were quantified by non-dispersive infrared detectors (NDIR) (accuracy of measuring cells: $\pm 2\%$ of full scale, scales: 0-20% for H₂ and CO, 0-10% for CO₂ and CH₄). In addition, a gas chromatograph (GC) equipped with two columns (using Ar as carrier gas) was used to measure the gas species concentrations and to specifically analyze the light hydrocarbons (C₂H_m). Three temperature sensors (Pt-Rh thermocouples) were settled to monitor the reactor temperatures (Figure 1). They were related to the inner temperature within the cavity volume (T₃ in the conical region near the cavity bottom and T₁ at the cavity center) and to the external wall of the cavity (T₂). The sensors tips were shielded by an alumina tube to protect them from the reacting gases. In addition, a solar-blind pyrometer (Impac, operating at 4.8–5.2 μm in a H₂O absorption band) placed above the reactor was used to measure the temperature at the cavity central core. Moreover, to ensure that the reactor was maintained at an operating pressure below 1 bar (atmospheric pressure of 860 hPa due to site elevation), a Venturi pump was placed at the outlet tube. A pressure measurement (P) was settled in the cavity to monitor pressure variations throughout the experiments. All the obtained data were controlled and collected by an automated data acquisition system (BECKHOFF).

2.3 Bed materials

Previous simulations using 3D CFD multiphysics modelling of the reactor (without bed materials) (Boujjat et al., 2019a,b) revealed that the cavity wall temperature was 100°C higher than the inner reaction zone temperature because of the direct absorption of most of the incoming radiations by the cavity wall. Moreover, the lightest particles were entrained early towards the outlet by the high velocity jet, which penalized the biomass conversion and syngas yields. Notable gradients in both steam concentration and gas temperature between the central spout region and the annular zone were also

pointed out by the model, leading to strong variations in the particles conversion rate. With the aim of achieving higher reactor performance and better use of solar energy, inert bed materials set in the form of spouted or packed bed were considered. The spouted flow regime is expected to increase the reaction zone temperature and enhance the gas/wood particles phase mixing. The packed-bed configuration is used to broaden the gas stream at the cavity bottom, which should reduce the central gas jet velocity (thus increasing the gas residence time) and limit the entrainment of the reacting particles, when compared with an empty cavity configuration. Figure SI2 provides an illustration of the three studied hydrodynamic configurations.

Table 3 presents the tested packed and spouted particles characteristics.

Table 3. Inert particles characteristics at 1400°C

	Alumina (Al ₂ O ₃)	Silicon carbide (SiC)	Olivine	Silica sand (SiO ₂)
Density, ρ_s (kg.m ⁻³)	3950	3210	3270	1442
Thermal conductivity, λ_s (W.m ⁻¹ .°C ⁻¹)	7	15	0.6	2.2
Heat capacity, $c_{p,s}$ (J.kg ⁻¹ .°C ⁻¹)	795	1320	1220	1225
Mean diameter, d_s (mm)	3.00 or 0.12	0.30	0.30	0.20

The olivine main compounds included MgO, SiO₂, CaO (0.4% max) and Fe₂O₃. The mass fractions in Mg, Si and Fe were respectively 28.5%, 17.5% and 7%. Both SiC and olivine particles were thermally treated at 1000°C in presence of air to ensure inertness regarding the biomass thermochemical reactions of pyrolysis and gasification. Figure 2 shows a photography of the different particles.

Figure 2. Studied inert bed and wood biomass particles

3. Modelling

Prior to the high temperature experiments, multiphysics simulations of three configurations (empty cavity, inert packed-bed and spouted-bed particles) were performed to analyze the gas/solid flow behavior under real processing conditions using the same bottom and upper (window) gas flow rates as experiments with a solar power input of 900W. The reactor geometry was elaborated to closely represent the real reactor design (composed of different zones: alumina cap, insulation, cavity walls, and fluid / gas injection, reaction and exit zones). The geometry was meshed using an optimal cell size to accurately solve the gradients in the central spout region of the reactor where sharp variations in terms of temperature, gas velocity, and species mole fraction occur (Boujjat et al., 2019a). Two thermal CFD models were developed. The first one simulated the fluid flow through the porous packed bed. In this model, the packed bed particles were modelled through a momentum sink, which was function of the permeability and the porosity of the medium. A blending between the energy of fluid and solid particles was considered for the governing transport equation determining the temperature in the packed bed region. The model also allowed tracking discrete char particles in space by solving in a Lagrangian reference frame the discrete particles momentum and energy equations. The second model used the Eulerian-Eulerian (granular) approach to simulate the directly irradiated spouted bed particles. The flow was therefore described by conservation equations written for each phase separately (gas and granular). The endothermal pyro-gasification reaction was modelled by a volumetric heat sink applied to the fluid region of the cavity. Momentum, energy and radiative intensity transport equations used in the modelling

of the packed and the spouted powders are provided in detail in Tables SI1-2. The reactor geometry, mesh and boundary conditions are also presented in the Supporting Information (Section 2.1).

4. Results and discussion

4.1 Hydrodynamic & thermal study

The empty and the packed-bed configurations were simulated without discrete particles injection. Figure 3 shows the temperature field inside the solar reactor for a solar power input of 900W. It was observed that for the two configurations, the cavity walls reached an average temperature of around 1230°C while the gas phase average temperature was around 930°C. The empty cavity was characterized by a central gas jet reaching a temperature of around 700°C at the center of the conical part of the cavity, which was relatively colder than the annular region. In the packed bed configuration, the gas phase temperature reached the same temperature (around 700°C) above the porous medium due to the cold window inlet Ar flow rate. The porous zone model allowed to simulate heat transfer through the porous packed bed (composed of 3mm particles). Accordingly, the bed top surface temperature was estimated at 1000°C at its center and approached 1100°C nearby the cavity walls. Due to the thermal resistance of the packed particles, and to the cooling of the bottom of the bed by the entering cold gases, the bed bottom temperature was lower (around 950°C).

Figure 3. Temperature distribution: empty vs. packed bed

The gas velocity magnitude field inside the reactor is presented in Figure 4.

Figure 4. Velocity magnitude field: empty vs. packed bed

The simulations revealed a significant attenuation of the inlet gas flow velocity when using the packed particles. The gas jet velocity at the cone entrance was about 8 m/s. It then decreased to less than 0.5 m/s just above the bed. The Ar flow from the top of the reactor flowed downward to hit the bed top surface with a velocity of around 0.3m/s. It then flowed with the inlet gases towards the outlet along the cavity walls. The steam distribution inside the reactor and the inlet gas (water steam+argon) pathlines for the two configurations are presented in Figure 5. In the empty configuration, the central cavity region exhibited the highest water steam concentration, whereas the packed bed broadened the inlet gas streamlines over the entire cavity section, suggesting improved homogenization of chemical kinetics in the whole cavity region.

Figure 5. Steam distribution and inlet gas streamlines

The spouted bed configuration was studied at cold and at real processing conditions. Prior to the CFD simulations, cold tests were carried out at ambient conditions with air on a transparent PMMA cavity mockup to visualize the mixing of the particles and determine the minimum spouting conditions. The obtained results for a 10g bed of SiC and Al_2O_3 particles are summarized in Figure 6.

Figure 6. Experimental cold tests on a replicated PMMA cavity mockup (10g of inert particles)

The bed minimum spouting velocity was determined by plotting the bed pressure drop as function of the inlet gas flow rate. Figures 6-a,b show the measured pressure drop vs. the air flow rate for a 10g bed of 300 μm SiC and 125 μm Al_2O_3 particles. It can be observed that by increasing the gas flow rate, the total pressure drop increased until reaching a maximum value that initiated the spouting of the particles (around 11 mbar for SiC and around 8 mbar for Al_2O_3 bed). Accordingly, minimum spouting flow rates of 1.5 NI/min ($v_{\text{exp,SiC}}=7.42$ m/s) and 0.16 NI/min ($v_{\text{exp,Al}_2\text{O}_3}=0.85$ m/s) were measured for the SiC and Al_2O_3 particles. In Figures 6-c to 6-f, a thin layer of wood particles was set at the top of the inert bed to visualize the stirring of the wood particles for $U/U_{\text{ms}} \sim 2$. A very stable spouting regime and well-ordered mixing of the particles was observed with the SiC particles, whereas the Al_2O_3 particles showed 3D instabilities that distorted the central gas jet path. As a result, the fountain height was constantly varying and changing its position, as illustrated in Figure 6-g. This undermined the particles recirculation from the top of the bed to the bottom. Such spouting instabilities were observed in previous studies (Olazar et al., 1992) and were due to different interdependent factors such as too large or too small cone angles. They could also occur when the height of the bed exceeded the maximum spoutable height, or when the spouted powders diameter was too small to be fluidized in a coherent and stable fashion.

These results were compared to 2D axisymmetric CFD simulations using the Eulerian approach. Figure 7 shows the spouted bed particles space distribution for different inlet gas flow rates.

Figure 7. Spouted bed particles space distribution for different inlet air flow rates (10g of inert particles, $t=2\text{s}$)

The cyclic particle-flow recirculation pattern through the three distinct regions (central spout, annular zone and fountain above the bed surface) was well reproduced by the Eulerian model. The predicted minimum spouting flow rate was 1.5 NI/min for the SiC bed ($v_{\text{sim,SiC}}=7.95$ m/s) and 0.23 NI/min for the

Al₂O₃ bed ($v_{sim,Al_2O_3}=1.22$ m/s). These predictions were in agreement with the experimentally measured values for SiC within an error range of 4% and slightly overestimated the minimum spouting flow rate by about 0.07 Nl/min for the Al₂O₃ particles. The observed discrepancies for the Al₂O₃ particles can be due to the aforementioned flow perturbations (Figure 6-g) that potentially facilitated the gas passage through the annular region. Such 3D instabilities could not be reproduced by the 2D axisymmetric model.

Subsequently, the reactor was simulated under real processing conditions (same bottom and upper gas flow rates as experiments) with a solar power input of 900W (and with 10g of spouted bed mass). The obtained temperature distribution is presented in Figure 8-a,b for Al₂O₃ and SiC spouted particles. Just as the two previous hydrodynamic configurations (empty and packed bed), the reactor cavity walls reached an average temperature of 1230°C. Unlike the empty configuration where the central gas jet reached only 700°C at the conical part of the reactor, the central gas jet when using the spouted particles reached high temperatures up to 1180°C right away.

Figure 8. Temperature distribution (gas and solid phases): (a) 125 μ m-Al₂O₃ particles; (b) 300 μ m-SiC particles

By comparing the simulations between Al₂O₃ and SiC, the alumina particles allowed increasing the gas phase temperature over a larger area. This is because the alumina particles were better spouted thanks to their lower diameter. Moreover, their temperature was more homogeneously distributed, whereas small gradients were noticed for SiC particles (Figure 8-b), arising from the less pronounced stirring of the SiC particles inside the cavity. This is confirmed by the different fountain heights depending on the particles diameter (Figure 9-b,d). Indeed, the fountain height of the Al₂O₃ bed was two to three times higher than the fountain height of the SiC particles. Moreover, the central gas flow (bottom jet zone) was more

attenuated by the SiC particles that somehow deflected part of the flow towards the annular zone (Figure 9-a,c). For readability reasons, the velocity magnitude field was plotted in a range of 0 m/s to 3 m/s. However, due to the heating of the gases inside the injection tube, the gas velocity increased from 3 m/s (at the inlet of the injection tube) to values above 9 m/s at the cone entrance for both 10g beds (Figure 10). This value is hence higher than the minimum spouting velocities (Figures 6 and 7-b); therefore, the spouting regime was largely achieved with the current experimental flow rates.

Figure 9. Gas velocity field and solid volume fraction: (a,b) 125 μ m- Al_2O_3 particles, (c,d) 300 μ m-SiC particles

The spouting gases (Ar+H₂O) pathlines colored by velocity magnitude are presented in Figure 10

Figure 10. Spouting gases pathlines colored by velocity magnitude

It clearly showed that for the Al_2O_3 particles, the flow crosses the bed mainly from its center to reach the fountain zone while the SiC spouted particles distribute part of the flow towards the annular region. Thereafter, the spouting gases interact with the upper window inlet flow to get routed all along the cavity walls towards the gas exit.

Overall, the simulations evidenced the benefits of using inert materials in the form of spouted particles to increase the reaction zone temperature while ensuring an efficient stirring of the particles. The experimental gas flow rates were high enough to spout the 10g bed. A slight increase of the inlet argon flow rate is recommended for the largest particles to better disperse them in the upper part of the cavity. Further increase of the spouting flow rate for the smallest bed particles can possibly impair the process by entraining them out of the conical cavity. The observed flow instabilities during cold tests for the Al_2O_3 spouted particles were not predicted by the 2D model. They are expected to negatively affect the heat transfer through the bed at high temperature.

A complementary comparative study of the three configurations based on hydrodynamic criteria only is provided in the Supporting Information, section 3.2.

4.2 Experimental study of solar reactor operation

Different experimental runs were performed to study the effect of the inert bed particles on biomass conversion and syngas composition. The Carbon Conversion Efficiency (CCE), the Cold Gas Efficiency (CGE) and the Solar-to-Fuel Efficiency (SFE) metrics were used to evaluate the performance of the reactor for each of the hydrodynamic configurations. The CCE quantifies the extent of biomass conversion inside the reactor. The CGE is expressed as the ratio between the calorific value of the syngas over that of the initial biomass. The SFE represents the ratio between the calorific value of syngas over the total thermal energy that enters the reactor including solar and biomass. For each gas species, the time-dependent production rate (Q_i) was assessed based on the measured species outlet mole fractions (x_i) and the inlet inert gas flow rate (Q_{Ar}): ($Q_i = Q_{\text{Ar}} \cdot x_i / x_{\text{Ar}}$). The averaged syngas mole composition (calculated over the experiment duration) was determined by integrating the gas production rates over time.

$$CCE = \frac{n_{\text{C in syngas}}}{n_{\text{C in biomass}}} \quad (1)$$

$$CGE = \frac{LHV_{\text{syngas}} \cdot m_{\text{syngas}}}{LHV_{\text{feedstock}} \cdot m_{\text{feedstock}}} \quad (2)$$

$$SFE = \frac{LHV_{\text{syngas}} \cdot m_{\text{syngas}}}{Q_{\text{solar}} + LHV_{\text{feedstock}} \cdot m_{\text{feedstock}}} \quad (3)$$

The tests were carried out under stable solar irradiation (DNI variation less than 10%). To stabilize the reactor temperature, the incident solar power was controlled by adjusting the opening of a trapdoor located between the heliostat and the reactor frame. The average solar power directly absorbed by the cavity through the aperture was $900 \pm 100\text{W}$ to maintain the reactor at 1200°C and $1200 \pm 100\text{W}$ to maintain the reactor at 1300°C .

The experimental conditions at 1200°C (Runs#1-7) and 1300°C (Runs#8-11) are recapped in Table 4. The calculated $(\text{S/B})/(\text{S/B})_{\text{st}}$ mole ratio was equal to 1.24 based on Eq.4 for calculating the $(\text{S/B})_{\text{st}}$ stoichiometric ratio (where S denotes steam flow rate including both inlet steam and moisture contained

in biomass, and B denotes dry biomass flow rate). Accordingly, the biomass particles were gasified with a slightly overstoichiometric steam flow rate.

Table 4. Operating conditions of the directly irradiated reactor (1200 and 1300 °C, 10g of inert materials)

Run#	T _{operating} (°C)	Bed material		Bed configuration	Wood feeding rate (g/min)	Ar flow rate (NL/min)	H ₂ O flow rate (g/min)
		material	size				
1	1200	Empty	-	Dilute spouted bed	1.2	0.2	0.2
2	1200	Al ₂ O ₃	3mm	Packed bed	1.2	0.2	0.2
3	1200	Al ₂ O ₃	125µm	Spouted bed	1.2	0.2	0.2
4	1200	SiC	300µm	Spouted bed	1.2	0.2	0.2
5	1200	SiC	300µm	Spouted bed	1.2	0.3	0.2
6	1200	Olivine	300µm	Spouted bed	1.2	0.2	0.2
7	1200	Sand	300µm	Spouted bed	1.2	0.2	0.2
8	1300	Al ₂ O ₃	3mm	Packed bed	1.2	0.2	0.2
9	1200-1300	Al ₂ O ₃	125µm	Spouted bed	1.2	0.2	0.2
10	1300	Al ₂ O ₃	125µm	Spouted bed	1.2	0.2	0.2
11	1300	SiC	300µm	Spouted bed	1.2	0.2	0.2

After solar tests, the material mass balance was assessed via systematic comparison between the initial mass of injected reactants (both biomass and water) and the output mass of products (syngas compounds, excess water and entrained char/soot/tar remaining in bubbler and filters). Overall, the mass balance closure was above 84% regardless of the experimental runs (as shown in Table SI3). Moreover, no remaining char was found inside the cavity after tests.

The reactor temperature as measured by T3 (inside the cavity) for Runs#1-11 is plotted in Figure 11-a,b. The temperature was effectively maintained around the set point values (maximum mean deviation of 14°C in Run#6). Due to the insufficient solar heating in Run#9, the reactor temperature could not be maintained at 1300°C (it declined to values approaching 1200°C) and the mean deviation was therefore higher (about 29°C).

Figure 11. T3 measurements for Runs#1-11 at 1200°C and 1300°C (left); Average and mean deviation of temperature (right)

Pyrometer measurements (T_{pyro}) in Figure 12 indicated strong fluctuations in temperature for the spouted-bed configuration because the pyrometer pointed directly at the bed top surface. Given the spouting of particles (including wood and inert particles) and the continuous feeding of fresh biomass, such fluctuations given by the optical pyrometer are expected (for simplification, the same emissivity of 1 was assumed for all the measurements, although in practice the emissivity is dependent on the materials properties and temperature, which also explains the observed differences in measured temperatures). The fluctuations, which reflect the intense/effective stirring of the particles, were lowered when the packed Al_2O_3 particles were used (Run#2).

Figure 12. Pyrometer measurements for Runs#1-7 (1200°C)

Figure 13 shows the calculated performance metrics and the gas composition for the different hydrodynamic configurations at 1200°C (Runs#1-7).

Figure 13. (a) CCE and top view of the cavity at the end of the experiments (for Al₂O₃, SiC and olivine); (b) Gas composition; (c) CGE; (d) SFE (Runs#1-7 at 1200°C)

The carbon conversion in the empty case (Run#1) reached 79.3% and the syngas produced was essentially composed of H₂ and CO. The H₂ and CO volume fractions were respectively 51.6% and

38.3%. The use of the packed and spouted bed of Al_2O_3 particles (Runs#2-3) increased the CCE up to 84.7%. In contrast, the CCE declined to 75.5% with the spouted 300- μm SiC (Run#4).

The increase of the spouting carrier gas flow rate (from 0.2 NL/min in Run#4 to 0.3 NL/min in Run#5) decreased even more the CCE to 73.9%. While the alumina particles remained intact at the end of the experiments and showed chemical inertness, the SiC bed suffered from sintering and adhered to the cavity walls (Figure 13-a). This can also be due to possible side reactions that may occur between SiC and water (forming either $\text{SiO}_2 + \text{CH}_4$ or $\text{SiO}(\text{g}) + 2\text{H}_2 + \text{CO}$, the former being the most thermodynamically favorable). The expected effect of improving the stirring of SiC particles by increasing the carrier gas flow rate was therefore hardly achieved and only impaired the gas and solid residence times.

The olivine sample in Run#6 also stuck but to a lesser extent on the cavity walls and its color turned from brown/red (due to the presence of iron oxides) to grey (Figure 13-a), suggesting poor chemical inertness at high temperatures. The CCE in Run#6 was still higher than the CCE of the empty cavity and reached 82.2%. The silica sand bed in Run#7 was the least resistant material, as the cavity was found almost empty at the end of the experiment, presumably because of the formation of fused silica during spouting caused by particles exposure to high-flux radiation in the upper cavity region. The results in terms of CCE and syngas composition were therefore very similar to Run#1.

In general, the bed materials only slightly influenced the gas composition and in all cases, the main syngas components remained H_2 and CO , whereas CO_2 , CH_4 and C_nH_m (mainly C_2H_2) were produced in smaller amounts. The Al_2O_3 particles in Run#2 led to the lowest H_2 concentration (~48.1%) while the light hydrocarbons ($\text{CH}_4 + \text{C}_n\text{H}_m$) volume fraction was the largest (6.4%). The H_2 concentration in Run#4 (SiC particles, 0.2 NL/min of spouting carrier gas flow rate) was the highest and reached around 52.9%. This however can be due to the oxidation of SiC by steam, forming additional H_2 in the mixture. The CGE (Figure 13-c) followed a similar pattern as the CCE since the gas composition was only slightly modified by the addition of the inert powders. The peak CGE values were obtained in Runs#2-3 (packed and spouted Al_2O_3 particles) and reached 1.13 against 1.11 in Run#1 (empty cavity). The minimum CGE was 1.01 in Run#5 (SiC bed) when the Ar flow rate was set to 0.3 NL/min.

The SFE (Figure 13-d) reached up to 21.8% in Run#1 (empty cavity) and decreased moderately to a minimum value of 19.4% in Run#2 (packed Al_2O_3 particles) even if the CGE was slightly upgraded. The total solar energy consumed (Q_{solar}) for gasifying the 30.0g biomass feedstock was 1.76 ± 0.21 MJ in Run#1. It was higher with inert bed materials: 2.23 ± 0.19 MJ in Run#2 (packed Al_2O_3 particles), 2.15 ± 0.24 MJ in Run#3 (spouted Al_2O_3 particles), 1.89 ± 0.21 MJ in Run#4 (spouted SiC particles), 1.83 ± 0.19 MJ in Run#5 (spouted SiC particles) and 2.04 ± 0.25 MJ in Run#6 (spouted olivine particles). Q_{solar} in Run#7 was 0.99 ± 0.11 MJ (because only 15.6g of biomass was gasified). While a small increase of Q_{solar} was observed when using the inert particles probably for heating the bed materials, the

uncertainty on Q_{solar} makes it difficult to precisely conclude about the influence of the bed materials on the solar energy consumption and therefore on the SFE.

The effect of temperature on the gasification process was studied in Runs#8-11 with the Al_2O_3 packed particles (Run#8), Al_2O_3 spouted bed particles (Runs#9-10) and with SiC bed (Run#11). Olivine and sand were not tested at this temperature, due to their low stability and resistance to the solar heating. Figure 14 shows the calculated reactor efficiencies and gas composition at 1300°C.

Figure 14. (a) CCE; (b) Gas composition; (c) CGE; (d) SFE (Runs#8-11 at 1300°C)

The temperature increase showed only a little impact on the CCE and CGE for the packed bed configuration (Run#8) but the influence on the gas composition was more pronounced. The CCE and the CGE both slightly declined by less than 1.5% whereas the syngas ($\text{H}_2 + \text{CO}$) concentration was improved by 4.3% (from Run#2 to Run#8) due to the enhancement of the kinetic rates of the steam methane and light hydrocarbons reforming ($\text{C}_n\text{H}_m + n\text{H}_2\text{O} \rightarrow (m/2+n) \text{H}_2 + n\text{CO}$).

The CCE in the Al_2O_3 spouted bed configuration declined with temperature increase, from 83.5% in Run#3 to 81.1% in Run#9 and 80.4% in Run#10. This decrease in CCE was ascribed to the more favored entrainment of the reacting char particles, caused by the lower gas residence time that decreased from 0.54s at 1200°C to 0.51s at 1300°C. Accordingly, the CGE declined in a stepwise fashion in Runs#9-10 to reach 109% in Run#10. Moreover, just as in Run#8, the $\text{H}_2 + \text{CO}$ volume fraction was improved by 3.2% at the expense of a noticeable decrease of the $\text{CH}_4 + \text{C}_n\text{H}_m$ by 28.0% (from Run#3 to Run#10). The largest $\text{H}_2 + \text{CO}$ volume fraction was achieved with the SiC particles (Run#11) reaching 94.2% against 92.1% in Run#4 (SiC bed-1200°C) with a beneficial effect on the CCE and the GGE that approached

values of the Al_2O_3 spouted bed possibly due to their better stirring and absorption of the solar flux at 1300°C.

For all runs (#8-11), the SFE decreased by more than 10% mainly due to the increase of the solar energy consumption. In fact, the calculated Q_{solar} for Runs#8-11 (1300°C) were higher than in Runs#1-7 (1200°C) and were respectively 2.49 ± 0.20 MJ, 2.68 ± 0.22 MJ, 2.25 ± 0.18 MJ, 2.16 ± 0.18 MJ.

5. Conclusions

This study focused on solar biomass gasification in a directly-irradiated conical lab-scale reactor and addressed the effect of inert particles on the gasification process. Prior to the experimental study, multiphysics CFD gas-particle flow modelling was developed to explore the impact of inert packed or spouted particles on the reactor thermal and hydrodynamic behavior.

(i) The empty conical cavity (no bed materials) is characterized by a high velocity central gas jet that entrains cyclically the char particles inside the cavity. It offers an excellent exposure to solar radiation evidenced by peak char temperature values but low gas residence time in the central cavity region due to high gas velocities.

(ii) The packed-bed configuration using inert particles at the bottom of the conical cavity significantly attenuates the gas jet speed at the central zone, broadens the gas streamlines and limits the entrainment of the reacting particles.

(iii) The spouted-bed configuration increases the reaction zone temperature and warrants a continuous gas/particles stirring. The finest inert spouted bed particles are more effective in increasing the reaction zone temperature as they are better spatially dispersed.

The experimental part of this study examined five different inert bed materials at 1200°C and 1300°C.

The empty configuration (no bed materials used), in which only the reacting wood particles are spouted, showed a CCE of 79% and a CGE of 111% at 1200°C. The Al_2O_3 packed and spouted particles both increased the reactor performance, especially the CCE that reached 85% in the packed bed configuration. The SiC bed suffered strongly from sintering and led to the lowest performance with a CCE of 76% and a CGE of 105%. Olivine and silica sand were not thermochemically stable at 1200°C, with global performance equivalent to the empty configuration. The Al_2O_3 beds were the most suitable for the high-temperature process showing good resistance to thermal shocks and chemical inertness. The effect of bed materials on the outlet syngas composition at these temperatures remained very low (less than 7% for H_2). Increasing temperature to 1300°C improved the syngas quality but globally impaired the CCE, CGE and SFE.

Future work should focus on the optimization of the bed particles properties. Low density and high absorption are preferable. New reactor configurations using a confiner could also limit particle entrainment that is a major concern for scale up.

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References

- Bellouard Q., Abanades S., Rodat S., Dupassieux N., 2017a. Solar thermochemical gasification of wood biomass for syngas production in a high-temperature continuously-fed tubular reactor. *Int. J. Hydrog. Energy* 42, 13486-13497.
- Bellouard Q., Abanades S., Rodat S., Dupassieux N., 2017b. Biomass Gasification in an Innovative Spouted-Bed Solar Reactor: Experimental Proof of Concept and Parametric Study. *Energy & Fuels* 31, 10933-10945.
- Bellouard Q., Abanades S., Rodat S., 2018. Design, simulation and experimental study of a directly-irradiated solar chemical reactor for hydrogen and syngas production from continuous solar-driven wood biomass gasification. *Int. J. Hydrog. Energy* 44, 19193-19205.
- Boujjat, H., Rodat, S., Chuayboon, S., Abanades, S., 2019a. Numerical simulation of reactive gas-particle flow in a solar jet spouted bed reactor for continuous biomass gasification. *Int. J. Heat Mass Transf.* 144, 118572.
- Boujjat, H., Rodat, S., Chuayboon, S., Abanades, S., 2019b. Experimental and numerical study of a directly irradiated hybrid solar/combustion spouted bed reactor for continuous steam gasification of biomass. *Energy* 189, 116118.
- Chuayboon, S., Abanades, S., Rodat, S., 2018a. Comprehensive performance assessment of a continuous solar-driven biomass gasifier. *Fuel Process. Tech.* 182, 1-14.
- Chuayboon, S., Abanades, S., Rodat, S., 2018b. Experimental analysis of continuous steam gasification of wood biomass for syngas production in a high-temperature particle-fed solar reactor. *Chem. Eng. & Proc.: Proc. Intens.* 125, 253-265.

513 Erkiaga, A., Lopez, G., Amutio, M., Bilbao, J., Olazar, M., 2013. Steam gasification of biomass in a
514 conical spouted bed reactor with olivine and γ -alumina as primary catalysts. *Fuel Process. Technol.* 116,
515 292–299.

516 Gokon, N., Ono, R., Hatamachi, T., Liuyun, L., Kim, H.J., Kodama, H.J., 2012. CO₂ gasification of coal
517 cokes using internally circulating fluidized bed reactor by concentrated Xe-light irradiation for solar
518 gasification. *Int. J. Hydrog. Energy* 37, 12128-12137

519 Gregg, G.W., Taylor, R.W., Campbell, J.H., Taylor, J.R., Cotton, A., 1980. Solar gasification of coal,
520 activated carbon, coke and coal and biomass mixtures. *Sol. Energy* 25, 353-364.

521 Hathaway, B.J., Kittelson, D.B., Davidson, J.H., 2014. Development of a Molten Salt Reactor for Solar
522 Gasification of Biomass. *Energy Procedia* 49, 1950–1959.

523 Higman, C., 2013. State of the Gasification Industry – the Updated Worldwide Gasification Database,
524 Germany.

525 Kodama T., Gokon, N., Enomoto, S., Itoh, S., Hatamachi, T., 2010. Coal Coke Gasification in a
526 Windowed Solar Chemical Reactor for Beam-Down Optics. *J. Sol. En. Eng.* 132(4):041004.

527 Kodama, T., Kondoh, Y., Tamagawa, T., Funatoh, A., Shimizu, K.I., Kitayama, Y., 2002. Experimental
528 investigation of a packed-bed solar reactor for the steam-gasification of carbonaceous feedstocks. *Energy*
529 & Fuels 16, 1264-1270.

530 Kruesi, M., Jovanovic, Z.R., Dos Santos, E.C., Yoon, H.C., Steinfeld, A., 2013. Solar-driven steam-based
531 gasification of sugarcane bagasse in a combined drop-tube and fixed-bed reactor – Thermodynamic,
532 kinetic, and experimental analyses. *Biomass & Bioenergy* 52, 173–183.

533 Li, X., Dai, Y.J., Wang, R.Z., 2015. Performance investigation on solar thermal conversion of a conical
534 cavity receiver employing a beam-down solar tower concentrator. *Sol. Energy* 114, 134–151.

535 Lopez, G., Cortazar, M., Alvarez, J., Amutio, M., Bilbao, J., Olazar, M., 2017. Assessment of a conical
536 spouted with an enhanced fountain bed for biomass gasification. *Fuel* 203, 825–831.

537 Loutzenhiser, P.G., Muroyama, A.P., 2017. A review of the state-of-the-art in solar-driven gasification
538 processes with carbonaceous materials, *Sol. Energy* 156, 93-100.

539 Makibar, J., Aranzazu, R.F-A., Diaz, L., Gartzen, L., Olazar, M., 2012. Pilot scale conical spouted bed
540 pyrolysis reactor: Draft tube selection and hydrodynamic performance. *Powder Technol.*, 219, 49-58.

541 Markowski, A., Kaminski, W., 1983. Hydrodynamic characteristics of jet-spouted beds. *The Canadian J.*
542 *of Chem. Eng.* 62, 377-381.

543 Mathur, K.B., Gishler, P.E., 1955. A technique for contacting gases with coarse solid particles. *AIChE J.*
544 1,157–164.

545 Müller, F., Poživil, P., Van Eyk, P.J., Villarrazo, A., Haueter, P., 2017. A pressurized high-flux solar
546 reactor for the efficient thermochemical gasification of carbonaceous feedstock. *Fuel* 193, 432–443.

547 Noone, C.J., Torrilhon, M., Mitsos, A., 2012. Heliostat field optimization: A new computationally
548 efficient model and biomimetic layout. *Sol. Energy* 86, 792–803.

549 Olazar, M., San Jose, M.J., Aguayo, A.T., Arandes, J.M., Bilbao, J., 1992. Stable operation conditions for
550 gas-solid contact regimes in conical spouted beds. *Ind. Eng. Chem. Res.* 31, 1784–1792.

551 Piatkowski, N., Wieckert, C., Steinfeld, A., 2009. Experimental investigation of a packed-bed solar
552 reactor for the steam-gasification of carbonaceous feedstocks. *Fuel Process. Technol.* 90, 360–366.

553 Puig-Arnabat, M., Tora, E.A., Bruno, J.C., Coronas, A., 2013. State of the art on reactor designs for solar
554 gasification of carbonaceous feedstock, *Sol. Energy* 97, 67–84.

555 Safarian, S., Unnpórsson, R., Richter, C., 2019. A review of biomass gasification modelling, *Renew.*
556 *Sustain. Energy Rev.* 110, 378–391.

557 Taylor, R.W., Berjoan, R., Coutures, J.P., 1983. Solar gasification of carbonaceous materials, *Sol. Energy*
558 30, 513–525.

559 Z'Graggen, A., Haueter, P., Trommer, D., Romero, M., De Jesus, J.C., Steinfeld, A., 2006. Hydrogen
560 production by steam-gasification of petroleum coke using concentrated solar power—II Reactor design,
561 testing, and modeling. *Int. J. Hydrog. Energy*, vol. 31, no. 6, pp. 797–811, 2006.

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