A pressurized high-flux solar reactor for the efficient thermochemical gasification of carbonaceous feedstock

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Abstract

We report on the design and first-ever experimental demonstration of a 3 kW pressurized solar reactor for thermochemically converting carbonaceous feedstocks into gaseous fuels. It uses a windowless SiC cavity to efficiently absorb and transfer concentrated solar radiation to an annular gas-particle vortex flow created by injecting tangentially a charcoal/water slurry at high pressures. Experiments were carried out in a high-flux solar simulator under a solar concentration ratio equivalent to 3718 suns. For slurry feeding rates in the range 0.42 – 1.26 g/min, H₂O:C molar ratios in the range 1.48 – 1.98, and absolute reactor pressures in the range 1 – 6 bar, the nominal reactor temperature was between 1009 and 1273 °C yielding high-quality syngas with a carbon conversion up to 94% within residence times of less than 5

seconds. The peak solar-to-fuel energy conversion efficiency, defined as the ratio of the heating value of the syngas produced to the solar radiative energy input plus the heating value of the slurry converted, reached 20%. The calorific value of the feedstock was solar upgraded by 35%, thus outperforming autothermal gasification in addition to delivering higher syngas output per unit of feedstock and eliminating the need for upstream air separation. The performance of this robust windowless design also was compared with that of an equivalent windowed configuration in which the gas-particle vortex flow was directly exposed to high-flux solar irradiation.

1. Introduction

Solar gasification thermochemically converts carbonaceous feedstocks into energy-rich and high-quality syngas – a mixture of CO and H₂ – using concentrated solar energy [1]. The thermochemical conversion involves principally two sequential processes: pyrolysis and steam-based char gasification, both of which are critical in the handling of carbonaceous fuels over a wide temperature range, with the former occurring typically in the range 200 – 600 °C and the latter becoming dominant at above 1000 °C. The simplified net reaction for stoichiometric water delivery can be represented by:

where x and y are the elemental molar ratios of H:C and O:C in the feedstock, respectively. Nitrogen, sulfur, mineral matter, and other impurities contained in the feedstock are omitted from consideration in Eq. (1). Both pyrolysis and gasification are highly endothermic. The advantages of the solar-driven process vis-à-vis the conventional autothermal process are four-fold: 1) it delivers higher syngas output per unit of feedstock, as no portion of the feedstock is combusted for process heat; 2) it produces syngas with higher calorific value and lower CO₂ intensity, as the energy content of the feedstock is upgraded through the solar

energy input; 3) it allows for higher gasification temperatures (>1200 °C), resulting in faster reaction kinetics and superior quality of the syngas produced with low – or without – tar content, resulting in better exploitation of the available resources; and 4) it eliminates the need for upstream air separation for oxy-combustion. Ultimately, solar gasification offers an efficient means of storing intermittent solar energy in a transportable and dispatchable chemical form. To overcome the intermittency of solar radiation, hybrid solar gasification concepts are proposed for around-the-clock process operation [2–5].

The measure of how well the solar energy input is converted into chemical energy contained in the syngas produced is given by the solar-to-fuel energy conversion efficiency $\eta_{\text{solar-to-fuel}}$, defined as:

$$\eta_{\text{solar-to-fuel}} = \frac{\dot{m}_{\text{syngas}} \cdot LHV_{\text{syngas}}}{\dot{Q}_{\text{solar}} + \dot{m}_{\text{feedstock}} \cdot LHV_{\text{feedstock}}} \quad (2)$$

where $\dot{Q}_{\text{solar}}$ is the solar energy delivered through the solar reactor's aperture, $\dot{m}_{\text{feedstock}}$ and $\dot{m}_{\text{syngas}}$ are the gasified feedstock and the evolved syngas mass rates, respectively, and LHV is the low heating value of the feedstock converted. Another important performance indicator is the energetic upgrade factor U , defined as the ratio of the heating value of the syngas produced to that of the feedstock processed:

$$U = \frac{\dot{m}_{\text{syngas}} \cdot LHV_{\text{syngas}}}{\dot{m}_{\text{feedstock}} \cdot LHV_{\text{feedstock}}} \quad (3)$$

For the stoichiometric system $\text{C} + \text{H}_2\text{O}$, the equilibrium composition at 1300 K consists of an equimolar mixture of H_2 and CO , yielding $U = 1.33$ for solar gasification [6]. Obviously, for autothermal gasification, $U < 1$, and typically $U = 0.75 - 0.85$ depending on the feedstock [7,8].

Solar gasification reactors may be classified [1] into: 1) directly-irradiated reactors, where the solid carbonaceous reactants are directly exposed to concentrated solar irradiation; and 2)

indirectly-irradiated reactors, where heat is transferred to the reaction site through an opaque wall. While directly-irradiated gasification reactors provide efficient heat transfer directly to the reaction site, they require a transparent window for the access of concentrated solar radiation, which becomes a critical and troublesome component at large scales and, particularly, under industrially-preferred high pressures. On the other hand, indirectly-irradiated reactors eliminate the need for a window at the expense of having less efficient heat transfer by conduction through the walls of an opaque absorber. Thus, the disadvantages are linked to the limitations imposed by the materials of the absorber with regard to maximum operating temperature, inertness to the chemical reaction, thermal conductivity, radiative absorptance, and resistance to thermal shocks. Previous experimental studies have used solar reactor concepts based on packed-beds [9–12], fluidized-beds [13–17], drop-tubes [18,19], and entrained flows [20,21]. Of particular interest is the directly-irradiated vortex flow reactor [20], which was successfully applied for the solar conversion of petcoke particles. However, its window became critical component during scale-up and pressurized operation because of the structural and thermal stresses [22]. This prompted us to consider converting the directly-irradiated configuration to an indirectly-irradiated one by replacing the window with a SiC cavity, since this configuration was recently applied successfully for heating pressurized air to 1200 °C at 6 bar using a 250 mm-inner diameter SiC cavity for a solar radiative power input of 50 kW [23].

The present paper describes the engineering design and experimental demonstration of a 3 kW indirectly-irradiated (windowless) vortex flow solar reactor for the thermochemical gasification of carbonaceous particles at high pressures. To our knowledge, this is the first-ever experimental demonstration of a pressurized solar gasifier. Its performance, in terms of the syngas quality, carbon conversion, upgrade factor, and solar-to-fuel energy conversion efficiency, is evaluated in the pressure range 1 – 6 bars, and compared to that of a directly-irradiated (windowed) vortex flow reactor.

2. Experimental

Solar Reactors – Two solar reactor configurations, shown schematically in Figure 1, were investigated: 1) the indirectly-irradiated (windowless) reactor, denoted I-I (Figure 1a); and 2) the directly-irradiated (windowed) reactor, denoted D-I (Figure 1b). Both the I-I and D-I solar reactors feature a gas-particle vortex flow following a helical path. Table 1 lists their key dimensions and materials. The I-I solar reactor consists of a flanged cylindrical SiC cavity, 30 mm-aperture, 95 mm-inner length, 40 mm-inner diameter, 5 mm-thick, with a hemispherical dome. This cavity serves as the solar absorber with an apparent absorptivity of 98.8% determined by Monte Carlo ray tracing. Graphite gaskets seal the flanges. A coaxial steel/Inconel 601 shell, lined with a $\text{SiO}_2\text{-Al}_2\text{O}_3$, creates an annular gap for the vortex flow. Concentrated solar radiation is absorbed by the SiC cavity, conducted through its walls, and transferred to the gas-particle vortex flow by combined conduction, convection, and radiation. To generate the vortex flow, Ar or CO_2 at a flow rate of up to $15 \text{ L}_\text{N}/\text{min}^\dagger$ was injected tangentially and directed toward the slurry feeding nozzle, as depicted in Figure 1a. The design of the D-I solar reactor was previously described in detail [20]; only the main features are highlighted here. The gas-particle vortex flow is confined to a 200 mm-length 100 mm-diameter cylindrical cavity-receiver, made of Inconel 601, and directly exposed to high-flux irradiation entering through a 50 mm-diameter aperture sealed with a 3 mm-thick transparent quartz disk. Two Ar flows were injected: one via a tangential nozzle directly behind the window at a rate of $2 \text{ L}_\text{N}/\text{min}$ to cool the window and protect it from particle deposition, and a second via four tangential nozzles behind the aperture at a rate of $3 \text{ L}_\text{N}/\text{min}$ to support the vortex flow.

[†] L_N denotes normal liters; mass flow rates calculated at 273.15 K and 1 atm.

Figure 1 - Schematic of: a) the indirectly-irradiated (I-I) solar reactor; and b) the directly-irradiated (D-I) solar reactor.

Table 1 - Key dimensions and materials of the I-I and the D-I solar reactors

Configuration:	I-I	D-I
Scheme:		
Thermocouple locations:	<p> $z1 = 25 \text{ mm}$ $z2 = 60 \text{ mm}$ $z3 = 110 \text{ mm}$ $r0 = 0 \text{ mm}, r1 = 25 \text{ mm},$ $r2 = 50 \text{ mm}$ </p>	<p> $z1 = 30 - 50 \text{ mm}$ $z2 = 110 - 135 \text{ mm}$ $z3 = 175 - 190 \text{ mm}$ $r2 = 50 \text{ mm}, r3 = 61 \text{ mm},$ $r4 = 80 \text{ mm}$ </p>
Absorber cavity:		
Material:	SiC	Al_2O_3
Material emissivity:	0.9	0.4

Apparent absorptivity:	0.988	0.93
Inner diameter:	40 mm	100 mm
Outer diameter:	50 mm	-
Inner length	95 mm	200 mm
Wall thickness	5 mm	-
Reactor chamber:		
Diameter:	100 mm	100 mm
Length:	110 mm	200 mm
Volume	0.8 l	1.6 l
Material:	Al ₂ O ₃ -SiO ₂	Al ₂ O ₃
Reactor shell:		
Material:	Stainless steel	Inconel 601
Diameter:	270 mm	120 mm
Thickness:	5 mm	1 mm
Length:	188 mm	260 mm
Front:		
Material:	Inconel 601	Aluminium
Aperture diameter:	30 mm	50 mm
Cooling:	Air-cooled	Water-cooled
Thermal insulation:		
Material:	Al ₂ O ₃ -SiO ₂	Al ₂ O ₃ -SiO ₂
Thickness:	78 mm	20 mm
Porosity:	64%	> 70%
Ceramic lining:		
Material:	SiO ₂ -Al ₂ O ₃	Al ₂ O ₃
Thickness:	10 mm	10 mm
Porosity:	< 30%	38%
Gas flow rates:		
Vortex flow:	2 - 15 L _N /min	3 L _N /min
Window protection:	-	2 L _N /min
Pressure rating:	1 – 6 bar	1.1 bar

Feedstock – Table 2 lists the ultimate (LECO CHNS- 932 & RO-479), and proximate (Netsch 409STA) analyses of activated charcoal (Fluka, Sigma-Aldrich 05120), which was used as the model carbonaceous feedstock. The particle size distribution, determined by laser scattering (HORIBA LA-950) was: $d_{\text{mean}} = 42.25 \mu\text{m}$ with $d_{v10} = 9.39 \mu\text{m}$, $d_{v50} = 24.63 \mu\text{m}$ (median), and $d_{v90} = 99.30 \mu\text{m}$. The lower heating value, determined based on the ultimate analysis [24], was $LHV_{\text{feedstock}} = 29.33 \text{ MJ/kg}$. A liquid slurry was prepared by mixing the charcoal with demineralized water to achieve a desired H₂O:C molar ratio. The slurry was injected at a desired charcoal feeding rate $\dot{m}_{\text{feedstock}}$ using a piston pump head (Ismatec QP Q2.CSC) and a peristaltic pump head (Ismatec MCP) for the I-I and D-I setups, respectively.

Table 2 - Ultimate (dry and ash free basis) and proximate analyses of activated charcoal (Fluka, Sigma-Aldrich 05120).

Element	wt. %	mole %
C	95.030	91.985
H	0.494	5.695
O	2.134	1.551
S	0.749	0.271
N	0.600	0.498
<hr/>		
Fixed carbon	90.16	-
Volatiles	2.37	-
Moisture	1.62	-
Ash	5.83	-

Experimental set-up – The experimental setup is shown in Figure 2. Experimentation was conducted at the high-flux solar simulator (HFSS) of ETH Zurich, which comprises an array of 7 high-pressure Xenon arcs, each close-coupled with truncated ellipsoidal specular reflectors, to provide an external source of intense thermal radiation – mostly in the visible and IR spectra – that closely approximates the heat transfer characteristics of highly concentrating solar systems [25]. The solar radiative input power $\dot{Q}_{\text{solar}}$ was measured optically on a Lambertian target with a calibrated CCD camera and verified with a water calorimeter. Temperatures were measured with type-K thermocouples at z - r - ϕ locations indicated in the schemes of Table 1. The nominal reactor temperature T_{reactor} , the nominal shell temperature T_{shell} , and the nominal cavity temperature T_{cavity} (I-I configuration only) were calculated as average of the corresponding local measurements. For the D-I configuration, a two-color pyrometer (OMEGA iR2) was employed to measure T_{reactor} immediately after the HFSS was turned off. A back-pressure valve (Tescom, 44-2500) was installed downstream and the pressure of the reactor chamber was monitored by a pressure transmitter (Keller, 33x). The gas mass flow rates were controlled by electronic flow controllers (Bronkhorst HI-TEC) calibrated for an accuracy of $\pm 1\%$. The composition of

gaseous products was analyzed on-line by infrared detectors (Siemens Calomat 6 and Ultramat; frequency 1 Hz) and by gas chromatography (Varian Micro-GC; frequency 8 mHz). A pre-set flow rate of N₂ (purity: 99.999%) was introduced into the product gas stream as tracer for the determination of the molar flow rates of species. During a typical experimental run, the system was firstly heated to the desired temperature under inert gas flow (Ar; purity of 99.996%) and then kept isothermally while feeding the reacting slurry. The main operational parameters were the solar radiative input power $\dot{Q}_{\text{solar}}$, the reactor's absolute pressure p , the charcoal feeding rate $\dot{m}_{\text{feedstock}}$, the H₂O:C molar ratio, and the Ar mass flow rate $\dot{m}_{\text{Ar}}$ supporting the vortex flow. In selected runs, Ar was substituted by CO₂ (purity: 99.998%) to study its effect on the gasification. The measurement errors were evaluated based on the standard deviation error propagation [26]. Main source of uncertainty was in the measurement of the mass flow rate of the slurry because of particle deposition in the feeding system.

Figure 2 - Solar experimental setup: (1) scale, (2) slurry piston pump, (3) mass flow controllers, (4) high-flux solar simulator, (5) spring-over pressure valve, (6) pressure transmitter, (7) condenser, (8) steam trap, (9) scrubber, (10) back-pressure valve, (11) gas analysis.

3. Results and Discussion

3.1 The I-I solar reactor

Table 3 lists the operating conditions and experimental results under approximately steady-state conditions for 51 solar experimental runs of the I-I solar reactor. The solar reactor was operated under the following range of parameters: $p = 1.0 - 5.9$ bar, $\dot{m}_{\text{Ar}} = 2 - 15$ L_N/min, $\dot{m}_{\text{feedstock}} = 0.42 - 1.26$ g/min, H₂O:C molar ratio = 1.48 – 1.98 (accounting for the moisture content in the charcoal), and $\dot{Q}_{\text{solar}} = 1.3 - 2.6$ kW, which corresponds to mean solar concentration ratios[‡] over the aperture in the range $C = 1794 - 3718$ suns. The highest T_{reactor} was 1273 °C, and T_{cavity} was 155 K higher on average. The highest operational pressure tested was 5.9 bar. Both T_{cavity} and p were mainly limited by the integrity of the SiC cavity. The nominal residence time of the reactants τ was calculated using [20]:

$$\tau = \frac{pV}{RT_{\text{reactor}}} \left(\dot{n}_{\text{Ar}} + \dot{n}_{\text{H}_2\text{O}} + \dot{n}_{\text{C}} \left(1 + \frac{X_{\text{C}}}{\ln(1 - X_{\text{C}})} \right) \right)^{-1} \quad (4)$$

where V is the volume of the reactor, R the universal gas constant, $\dot{n}$ the molar flow rate, and X_{C} the final extent of carbon conversion,

$$X_{\text{C}} = \frac{\dot{n}_{\text{CO}} + \dot{n}_{\text{CO}_2}}{\dot{n}_{\text{C}}} \quad (5)$$

For all runs of Table 3, τ was in the range 0.7 – 4.6 s. Note that since $X_{\text{C}} < 1$ (incomplete conversion), only the converted feedstock was accounted in Eqs. (2) and (3). The peak values of the performance indicators obtained were: $X_{\text{C}} = 94.4\%$, $\eta_{\text{solar-to-fuel}} = 19.7\%$, and $U = 1.35$.

[‡] The solar concentration ratio C is defined as the mean solar radiative flux over the solar reactor's aperture, normalized to the direct normal solar irradiation.

Table 3 - Operating conditions for the I-I solar reactor and experimentally measured key parameters under approximately steady-state conditions.

#	$\dot{Q}_{\text{solar}}$ [W]	p [bar]	$\dot{m}_{\text{Ar}}$ [L _N /min]	$\dot{m}_{\text{feedstock}}$ [g/min]	H ₂ O:C	T_{reactor} [°C]	T_{cavity} [°C]	$\dot{n}_{\text{H}_2}$ [mmol/min]	$\dot{n}_{\text{CO}}$ [mmol/min]	$\dot{n}_{\text{CO}_2}$ [mmol/min]	τ [s]	X_{C} [%]	$\eta_{\text{solar-to-fuel}}$ [%]	U
1	1546	1.0	4.0	0.48	1.48	1105	1248	19.4	12.6	3.4	1.9	45.2	8.3	1.20
2	1268	1.0	4.0	0.48	1.48	1010	1159	12.8	11.0	0.9	2.0	34.2	7.7	1.18
3	1971	1.0	4.0	0.50	1.48	1196	1351	35.7	29.1	3.3	1.6	89.2	12.9	1.23
4	1891	1.0	4.0	0.68	1.48	1185	1333	23.5	14.1	4.7	1.6	37.9	8.0	1.22
5	1934	1.0	4.0	0.91	1.48	1195	1338	38.4	24.7	6.9	1.4	47.5	12.5	1.21
6	1934	1.0	7.5	0.91	1.48	1191	1338	30.8	19.1	5.8	0.9	37.6	10.1	1.23
7	1934	1.0	11.0	0.93	1.48	1184	1331	27.9	18.3	4.8	0.7	34.0	9.4	1.20
8	1915	1.0	2.0	0.88	1.48	1204	1351	41.0	26.0	7.5	2.0	51.8	13.5	1.17
9	1671	1.0	4.0	0.62	1.73	1090	1218	25.8	18.2	3.8	1.7	48.6	10.5	1.21
10	2085	1.0	4.0	0.56	1.73	1214	1378	41.6	29.1	6.2	1.5	86.8	13.0	1.21
11	2238	1.0	4.0	0.56	1.73	1270	1439	39.6	27.1	6.2	1.5	81.9	11.4	1.20
12	2238	1.0	4.0	1.12	1.73	1256	1324	76.2	46.8	14.7	1.1	74.8	19.7	1.18
13	2154	1.0	4.0	0.85	1.73	1255	1418	50.8	31.5	9.6	1.3	66.1	14.3	1.18
14	1965	1.0	4.0	0.80	1.73	1202	1366	45.0	26.5	9.3	1.4	61.2	13.7	1.18
15	1965	1.0	5.0	0.85	1.73	1199	1364	43.9	26.1	8.9	1.2	56.5	13.5	1.20
16	1965	1.0	6.0	0.90	1.73	1197	1268	39.8	23.4	8.2	1.0	47.9	12.3	1.22
17	1965	1.0	3.0	0.77	1.73	1202	1365	48.3	28.8	9.8	1.6	68.1	14.9	1.16
18	1965	1.0	2.0	0.77	1.73	1202	1365	48.5	28.6	9.9	2.0	68.3	14.8	1.17
19	1649	1.0	4.0	0.81	1.73	1118	1270	40.2	27.2	6.5	1.5	56.5	15.4	1.21
20	1411	1.0	4.0	0.82	1.73	1035	1189	27.5	22.6	2.5	1.6	41.6	13.7	1.20
21	1316	1.0	4.0	0.70	1.73	1030	1192	22.1	18.5	1.8	1.7	39.3	12.0	1.19
22	1737	1.0	4.0	0.62	1.73	1149	1305	41.1	31.4	4.9	1.5	79.3	15.8	1.23
23	1465	1.0	4.0	0.69	1.98	1022	1117	18.1	14.6	1.8	1.6	32.1	9.0	1.16
24	1465	1.0	4.0	0.42	1.98	1039	1133	20.8	16.9	2.0	1.9	61.2	10.3	1.19
25	1909	1.0	4.0	0.54	1.98	1186	1297	41.9	29.7	6.0	1.5	89.9	14.1	1.20
26	2245	1.0	4.0	0.54	1.98	1273	1405	41.8	30.4	5.7	1.4	90.7	12.1	1.22
27	1949	1.9	4.0	0.99	1.73	1009	1168	14.4	8.7	2.8	3.0	16.0	4.9	1.22
28	2397	1.9	4.0	0.83	1.73	1199	1360	68.0	38.0	15.0	2.4	86.8	16.0	1.19
29	1885	1.9	4.0	0.96	1.73	1088	1211	31.9	16.5	6.8	2.7	33.3	10.1	1.31
30	2026	1.9	4.0	0.96	1.73	1086	1205	50.4	30.3	10.0	2.6	57.8	15.8	1.22
31	2022	3.1	4.0	0.87	1.73	1112	1240	18.5	9.4	3.4	4.6	20.1	5.6	1.35
32	2245	3.1	4.0	0.83	1.73	1142	1381	40.1	24.5	7.0	4.4	51.7	11.5	1.31
33	2524	3.0	4.0	0.83	1.73	1166	1333	48.9	30.6	9.0	4.1	65.2	12.0	1.28
34	2385	1.0	8.0	0.90	1.73	1132	1334	36.4	21.1	7.4	0.9	43.0	9.5	1.28
35	2453	2.1	8.0	0.99	1.73	1153	1346	48.6	25.0	11.3	1.7	50.0	11.6	1.28
36	2453	3.1	8.0	0.97	1.73	1162	1348	52.1	26.9	12.0	2.6	54.6	12.4	1.28
37	2453	3.9	8.0	0.99	1.73	1167	1348	51.0	26.2	11.6	3.3	51.9	12.1	1.29
38	2453	5.0	8.0	0.83	1.73	1180	1351	35.8	14.7	9.3	4.3	39.5	8.3	1.31
39	2219	1.0	3.0	0.63	1.73	1173	1342	31.4	16.5	7.5	1.9	52.2	8.7	1.18
40	2261	1.0	3.0	0.78	1.73	1161	1327	40.1	21.3	9.4	1.7	54.1	10.9	1.23

41	2290	2.0	6.0	0.74	1.73	1158	1323	40.0	21.4	8.9	2.2	56.4	10.7	1.28
42	2400	3.0	9.0	0.66	1.73	1179	1341	42.6	23.9	8.7	2.5	67.1	11.1	1.30
43	2400	3.9	12.0	0.63	1.73	1178	1333	45.3	28.3	8.3	2.6	79.0	11.9	1.28
44	2400	4.9	15.0	0.67	1.73	1169	1322	45.7	29.1	8.3	2.6	75.9	12.1	1.27
45	2400	5.9	15.0	0.65	1.73	1164	1314	43.2	27.2	8.0	3.2	74.3	11.5	1.26
46	2400	4.0	12.0	0.93	1.73	1167	1322	60.9	36.9	11.6	2.5	71.1	15.4	1.28
47	2400	4.0	12.0	1.26	1.73	1158	1314	65.2	37.2	13.7	2.3	55.2	15.3	1.26
48	2346	1.0	3.0*	0.77	1.73	1157	1342	16.7	45.5	122.1	1.7	59.3	10.9	1.27
49	2346	2.0	6.0*	0.77	1.73	1135	1332	10.2	56.9	245.9	2.2	61.3	12.0	1.33
50	2448	3.0	9.0*	0.77	1.73	1119	1317	8.0	69.0	373.8	2.5	72.1	13.2	1.31
51	2628	3.9	12.0*	0.77	1.73	1120	1314	6.6	78.8	510.6	2.5	94.4	13.4	1.18

* Ar substituted by CO₂

Figure 3a presents a representative experimental run conducted with the I-I solar reactor at ambient pressure. Plotted are the temporal variations of $\dot{m}_{\text{feedstock}}$, $\dot{Q}_{\text{solar}}$, T_{cavity} , T_{reactor} , T_{offgas} , and the product gas evolution. The period of the experiment considered to approximate steady-state conditions is indicated by the two vertical dotted lines. For this run, these conditions were: $p = 1$ bar, $\dot{m}_{\text{Ar}} = 4$ L_N/min, $\dot{m}_{\text{feedstock}} = 0.54$ g/min, H₂O:C = 1.98, and $\dot{Q}_{\text{solar}} = 1.9$ kW, leading to $T_{\text{cavity}} = 1297$ °C, $T_{\text{reactor}} = 1186$ °C, $T_{\text{offgas}} = 953$ °C, and yielding a syngas flow of $\dot{n}_{\text{H}_2} = 41.9$ mmol/min, $\dot{n}_{\text{CO}} = 29.7$ mmol/min, and $\dot{n}_{\text{CO}_2} = 6$ mmol/min. The performance indicators obtained were: $X_{\text{C}} = 89.9\%$ within $\tau = 1.5$ s, $\eta_{\text{solar-to-fuel}} = 14.1\%$, and $U = 1.20$.

Figure 3 - Representative solar experimental run conducted at $p = 1$ bar with: a) the I-I solar reactor; b) the D-I solar reactor.

Figure 4 presents $\dot{n}$ for the species H_2 (■), CO (▲), and CO_2 (●) as a function of T_{reactor} at $p = 1.0$ bar and $\dot{m}_{\text{Ar}} = 4 \text{ L}_\text{N}/\text{min}$, and for three slurry compositions: a) $\dot{m}_{\text{feedstock}} = 0.49 \pm 0.02 \text{ g/min}$, $\text{H}_2\text{O}:\text{C} = 1.48$; b) $\dot{m}_{\text{feedstock}} = 0.61 \pm 0.05 \text{ g/min}$, $\text{H}_2\text{O}:\text{C} = 1.73$; and c) $\dot{m}_{\text{feedstock}} = 0.55 \pm 0.10 \text{ g/min}$, $\text{H}_2\text{O}:\text{C} = 1.98$, (Figure 4 a, b, c respectively). High-quality syngas containing low molar fraction of CO_2 was obtained, with $\dot{n}_{\text{syngas}}$ increasing with T_{reactor} . The presence of CH_4 was found to be negligible ($\dot{n}_{\text{CH}_4} < 1.19 \text{ mmol/min}$) and detected only in 5 out of 51 experimental runs, partly because of the low volatile fraction in the charcoal as CH_4 is typically generated during pyrolysis [27]. In a representative sample retrieved from the off-gas system, no tars were detected, which was attributed to the de-volatilized charcoal and the high gasification temperatures.

Figure 4 - Molar flow rate for the product gas species H_2 (■), CO (▲), and CO_2 (●) for three slurry compositions (a, b, c) as a function of T_{reactor} , obtained with the I-I solar reactor at $p = 1$ bar.

Figure 5 presents: X_{C} (a), $\eta_{\text{solar-to-fuel}}$ (b), U (c), and $\text{H}_2:\text{CO}$ molar ratio of the syngas produced (d) as a function of T_{reactor} for $p = 1.0$ bar and $\dot{m}_{\text{Ar}} = 4$ L_N/min, and for three slurry compositions: $\dot{m}_{\text{feedstock}} = 0.49 \pm 0.02$ g/min with $\text{H}_2\text{O}:\text{C} = 1.48$ (■); $\dot{m}_{\text{feedstock}} = 0.61 \pm 0.05$ g/min with $\text{H}_2\text{O}:\text{C} = 1.73$ (▲); and $\dot{m}_{\text{feedstock}} = 0.55 \pm 0.10$ g/min with $\text{H}_2\text{O}:\text{C} = 1.98$ (●). The reaction extent increases monotonically with T_{reactor} and leveled at 90.7% for $T_{\text{reactor}} > 1180$ °C. The dependence of X_{C} on $\text{H}_2\text{O}:\text{C}$ stoichiometry is inconclusive, partly because of competing effects of higher $p_{\text{H}_2\text{O}}$ leading to faster reaction rates versus shorter τ as the $\text{H}_2\text{O}:\text{C}$ molar ratio is increased. Excess steam also leads to a higher sensible heat sink. Similarly, a

peak in $\eta_{\text{solar-to-fuel}}$ at 15.8% for 1150 °C can be explained by the increase in X_C with T_{reactor} at the expense of higher conductive and radiative heat losses. Occasional clogging of the feeding system was observed with the $\text{H}_2\text{O}:\text{C}$ molar ratio of 1.48, and thus experimentation was continued only with the higher $\text{H}_2\text{O}:\text{C}$ molar ratios. The $\text{H}_2:\text{CO}$ molar ratio varied within the range 1.16 – 1.54 with T_{reactor} , presumably as a result of the off-gas stream undergoing the water-gas shift reaction at the exit of the solar reactor prior to sampling [28]. The averaged value of U for this set of experiments was found to be 1.20 ± 0.06 . Note that, in contrast to X_C , U is less sensitive to T_{reactor} because it considers only the calorific upgrade of the converted feedstock.

Figure 5 - a) Reaction extent, b) solar-to-fuel energy conversion efficiency, c) energetic upgrade factor, and d) $\text{H}_2:\text{CO}$ molar ratio as a function of T_{reactor} for three slurry compositions: $\dot{m}_{\text{feedstock}} = 0.49 \pm 0.02 \text{ g/min}$ with $\text{H}_2\text{O}:\text{C} = 1.48$ (■); $\dot{m}_{\text{feedstock}} = 0.61 \pm 0.05 \text{ g/min}$ with $\text{H}_2\text{O}:\text{C} = 1.73$ (▲); $\dot{m}_{\text{feedstock}} = 0.55 \pm 0.10 \text{ g/min}$ with $\text{H}_2\text{O}:\text{C} = 1.98$ (●), obtained with the I-I solar reactor at $p = 1 \text{ bar}$.

The input value of $\dot{m}_{\text{Ar}}$ was varied to control τ and $p_{\text{H}_2\text{O}}$. Figure 6 presents X_C and $\eta_{\text{solar-to-fuel}}$ as a function of τ and $p_{\text{H}_2\text{O}}$ for $\dot{m}_{\text{Ar}} = 11.0, 7.5, 4.0, 2.0 \text{ L}_\text{N}/\text{min}$ ((a) to (d), respectively). The experimental conditions were: $p = 1.0 \text{ bar}$, $\dot{m}_{\text{feedstock}} = 0.91 \pm 0.05 \text{ g/min}$, $\text{H}_2\text{O}:\text{C} = 1.48$, and

$T_{\text{reactor}} = 1193 \pm 10$ °C. The corresponding Ar inlet velocity $u_{\text{Ar,i}} = 25.4, 17.1, 9.0, 4.7$ m/s provided a qualitative measure for particle entrainment by the vortex flow since the drag force acting on a particle is proportional to $u_{\text{Ar,i}}^2$. As $\dot{m}_{\text{Ar}}$ was lowered from 11 to 2 L_N/min, τ and $p_{\text{H}_2\text{O}}$ increased leading to higher values of X_{C} , but approaching a constant value because of insufficient particle entrainment (particle drop-out) for $u_{\text{Ar,i}} < 10$ m/s, as verified by particle deposition inside the reaction chamber. It can be seen that $\eta_{\text{solar-to-fuel}}$ follows the same trend because of higher reaction extent and less energy wasted in heating Ar.

Figure 6 - Reaction extent and solar-to-fuel energy conversion efficiency as a function of nominal residence time and $p_{\text{H}_2\text{O}}$ for $\dot{m}_{\text{Ar}} = 11.0, 7.5, 4.0, 2.0$ L_N/min ((a) to (d), respectively), obtained with the I-I solar reactor. Experimental conditions: $p = 1.0$ bar, $\dot{m}_{\text{feedstock}} = 0.91 \pm 0.05$ g/min, $\text{H}_2\text{O}:\text{C} = 1.48$, $T_{\text{reactor}} = 1193 \pm 10$ °C.

Figure 7 presents X_{C} as a function of T_{reactor} for $\dot{m}_{\text{Ar}} = 4$ L_N/min, $\dot{m}_{\text{feedstock}} = 0.87 \pm 0.05$ g/min, and $\text{H}_2\text{O}:\text{C} = 1.73$ and for three pressure levels: $p = 1.0, 1.9$, and 3.1 bar. For all 3 pressures, X_{C} monotonically increases with T_{reactor} in a manner that is approximately linear over the measured range. With increasing p , the fit becomes steeper in accordance with Le Chatelier's principle for Eq. (1), i.e., higher pressures shift the thermodynamic equilibrium composition

to higher temperatures. The intersection around $T_{\text{reactor}} = 1120$ °C points out to non-linear competing effects as X_C either decreases or increases with pressure at above or below that temperature, presumably due to change in the swirl number which affects the actual residence time [29].

Figure 7 - Reaction extent as a function of T_{reactor} for $p = 1.0$ bar (■), 1.9 bar (▲), and 3.1 bar (●), obtained with the I-I solar reactor. Experimental conditions: $\dot{m}_{\text{Ar}} = 4$ L_N/min, $\dot{m}_{\text{feedstock}} = 0.87 \pm 0.05$ g/min, and $\text{H}_2\text{O}:\text{C} = 1.73$.

Figure 8 presents X_C (a), $\eta_{\text{solar-to-fuel}}$ (b), U (c), τ (d), $\text{H}_2:\text{CO}$ molar ratio (e), and $\text{CO}_2:\text{CO}$ molar ratio (f) are plotted as function of p in the range 1 – 6 bar and for three different vortex flow settings: $\dot{m}_{\text{Ar}} = 8$ L_N/min (▲); $\dot{m}_{\text{Ar,gi}} = 3, 6, 9, 12, 15, 15$ L_N/min for $p = 1, 2, 3, 4, 5, 6$ bar, respectively (●); and $\dot{m}_{\text{CO}_2,\text{gi}} = 3, 6, 9, 12$ L_N/min for $p = 1, 2, 3, 4$ bar, respectively (■). For these runs, $\dot{m}_{\text{feedstock}} = 0.79 \pm 0.07$ g/min, $\text{H}_2\text{O}:\text{C} = 1.73$, and T_{reactor} was kept in the range 1133 – 1168 °C. For the first set of experiments (▲), $\dot{m}_{\text{Ar}}$ was kept constant over the tested pressure range, leading to a linear increase of τ and $p_{\text{H}_2\text{O}}$ and a reduction in $u_{\text{Ar,i}}$ with p . As expected, X_C increases with p within the range 1 – 3 bar due to longer τ and higher $p_{\text{H}_2\text{O}}$. For $p > 3$ bar, the effect of the slower $u_{\text{Ar,i}}$ became predominant and X_C decreases because of insufficient particle entrainment, as verified by the presence of residual particles deposited inside the reaction

chamber. $\eta_{\text{solar-to-fuel}}$ follows the same trend as X_C . In the following two sets of experiments, $\dot{m}_{\text{Ar/CO}_2}$ was gradually increased with p in order to eliminate deposition. In the second set of experiments (●), X_C increases with p , but converges toward a constant value for $p > 4$ bar because of the less favorable thermodynamics. As it can be seen, $\eta_{\text{solar-to-fuel}}$ was less dependent on p due to the higher sensible heat sink of the additionally injected Ar. In the third set of experiments (■), Ar was substituted by CO₂ to eliminate the use of an inert gas and study the effect on the H₂:CO molar ratio. The calculation of X_C , Eq. (5), was adjusted to account for the injected CO₂. Also here, X_C increases with p because of longer τ and higher $p_{\text{H}_2\text{O}} + p_{\text{CO}_2}$, peaking at 94.4% for $p = 4$ bar. Despite the increasing sensible heat losses, overall $\eta_{\text{solar-to-fuel}}$ improves mainly because of the higher reaction extent. The mean temperature of the metallic shell increases on average by 7.8 K/bar and 17.3 K/bar for $\dot{m}_{\text{Ar}}$ and $\dot{m}_{\text{CO}_2}$, respectively, because of higher effective thermal conductivity of the porous thermal insulation. The drop in U when feeding $\dot{m}_{\text{CO}_2,\text{gi}}$ at $p = 4$ bar is attributed to competing reactions mechanisms by H₂O and CO₂, including adsorption/desorption processes at active sites [15,30] as the absolute amounts of H₂ produced decrease with p_{CO_2} (see Table 3, Exp. 48-51), detrimentally affecting the calorific value of the syngas despite the increase in X_C . For all sets of experiments $U > 1.18$, peaking for the run with CO₂ (■) at 1.33 for $p = 2$ bar. The molar ratios H₂:CO and CO₂:CO are independent of p for the sets with $\dot{m}_{\text{Ar}}$ (▲ & ●). Compared with conventional autothermal gasification, the syngas quality is significantly superior and suitable for direct use in FT synthetization [31]. For the run with CO₂ (■), H₂:CO decreases and CO₂:CO increases by one order of magnitude.

Figure 8 - a) Reaction extent, b) solar-to-fuel energy conversion efficiency, c) energetic upgrade factor, d) nominal residence time, e) $\text{H}_2:\text{CO}$ molar ratio, and f) $\text{CO}_2:\text{CO}$ molar ratio as function of p obtained with the I-I solar reactor for three different vortex flow settings: $\dot{m}_{\text{Ar}} = 8 \text{ L}_\text{N}/\text{min}$ ($\blacktriangle$); $\dot{m}_{\text{Ar,gi}} = 3, 6, 9, 12, 15, 15 \text{ L}_\text{N}/\text{min}$ for $p = 1, 2, 3, 4, 5, 6$ bar, respectively ($\bullet$); and $\dot{m}_{\text{CO}_2,\text{gi}} = 3, 6, 9, 12 \text{ L}_\text{N}/\text{min}$ for $p = 1, 2, 3, 4$ bar, respectively ($\blacksquare$). Additional experimental conditions: $\dot{m}_{\text{feedstock}} = 0.79 \pm 0.07 \text{ g}/\text{min}$, $\text{H}_2\text{O}:\text{C} = 1.73$, and $T_{\text{reactor}} = 1133 - 1168 \text{ }^\circ\text{C}$.

An energy balance was performed for each of the 51 experimental runs of Table 3. Considered were: convective and radiative heat losses from the metallic shell to ambient $\dot{Q}_{\text{convection,shell}}$ and $\dot{Q}_{\text{radiation,shell}}$, respectively, conductive heat loss to the air-cooled front $\dot{Q}_{\text{conduction,front}}$, convective heat loss from the absorber cavity $\dot{Q}_{\text{convection,cavity}}$, re-radiation through the aperture $\dot{Q}_{\text{reradiation}}$, reflection by the cavity $\dot{Q}_{\text{reflection}}$, sensible heating of reactants $\dot{Q}_{\text{sensible}}$, and heat of the reaction $\dot{Q}_{\text{chemical}}$. $\dot{Q}_{\text{convection,shell}}$ and $\dot{Q}_{\text{convection,cavity}}$ were calculated

using Nusselt correlations [32,33]. $\dot{Q}_{\text{conduction,front}}$ was calculated by applying Fourier's law using material properties provided by the manufacturers and from literature [34]. $\dot{Q}_{\text{radiation,shell}}$ and $\dot{Q}_{\text{reradiation}}$ were calculated using the radiosity method. $\dot{Q}_{\text{reflection}}$ was computed by Monte Carlo ray-tracing. $\dot{Q}_{\text{sensible}}$ and $\dot{Q}_{\text{chemical}}$ were calculated using the enthalpy changes for the species: Ar, charcoal, H₂O, H₂, CO, and CO₂ (NIST Janaf).

On average for all 51 experimental runs of Table 3: $\dot{Q}_{\text{convection,shell}} = 44.8\%$ and $\dot{Q}_{\text{radiation,shell}} = 8.4\%$ of $\dot{Q}_{\text{solar}}$, which can be reduced by scaling up and improving thermal insulation. Scaling the reactor diameter by a factor of 10 could potentially double $\eta_{\text{solar-to-fuel}}$, as the increased volume-to-surface ratio inherently reduces $\dot{Q}_{\text{convection,shell}}/\dot{Q}_{\text{solar}}$ and $\dot{Q}_{\text{radiation,shell}}/\dot{Q}_{\text{solar}}$ [35]. $\dot{Q}_{\text{conduction,front}} = 12.4\%$ of $\dot{Q}_{\text{solar}}$, which can in principle be reduced by improving the thermal insulation. $\dot{Q}_{\text{convection,cavity}} = 7.8\%$ of $\dot{Q}_{\text{solar}}$, which is difficult to reduce and will eventually increase with forced convection [36]. $\dot{Q}_{\text{reradiation}} = 13.0\%$ of $\dot{Q}_{\text{solar}}$, which can be further reduced by incorporating a secondary concentrator [37], enabling a smaller aperture for the same radiative power input and thereby also reducing $\dot{Q}_{\text{convection,cavity}}$. $\dot{Q}_{\text{reflection}} = 1.2\%$ of $\dot{Q}_{\text{solar}}$, which corresponds to the apparent absorptivity of the cavity approaching a blackbody absorber. $\dot{Q}_{\text{sensible}} = 9.7\%$ of $\dot{Q}_{\text{solar}}$, which can be minimized by feeding the reactants at the stoichiometric ratio and by avoiding inert gas use, and thus eliminating heating unreacted flows. Finally, $\dot{Q}_{\text{chemical}} = 2.7\%$ of $\dot{Q}_{\text{solar}}$ (peak value of 5.1%), which only accounts for the standard enthalpy change of Eq. (1) multiplied by X_C .

3.2 Directly-irradiated solar reactor

Table 4 lists the operating conditions and experimental results under approximately steady-state conditions for 30 experimental runs of the D-I solar reactor. The solar reactor was operated under the following range of parameters: $p = 1$ bar, $\dot{m}_{\text{Ar}} = 3$ L_N/min, $\dot{m}_{\text{feedstock}} = 0.63 - 2.03$ g/min, H₂O:C molar ratio = 3.24, and $\dot{Q}_{\text{solar}} = 2.4 - 3.6$ kW, which corresponds to $C = 1228 - 1816$ suns. For all runs, the reactor was operated at ambient pressure because of the

flat quartz window (196 mm-diameter, 3 mm-thickness), which can only withstand up to 0.1 bar pressure difference. A concave window would permit operating at higher pressures [38,39]. The nominal residence time was in the range 1.0 – 2.1 s. The peak values of the performance indicators obtained were: $X_C = 69.1\%$, $\eta_{\text{solar-to-fuel}} = 16.9\%$, and $U = 1.25$.

Table 4 - Operating conditions for the D-I solar reactor, with experimentally measured key parameters under approximately steady-state conditions.

#	$\dot{Q}_{\text{solar}}$ [W]	$\dot{m}_{\text{feedstock}}$ [g/min]	T_{reactor} [°C]	$\dot{n}_{\text{H}_2}$ [mmol/min]	$\dot{n}_{\text{CO}}$ [mmol/min]	$\dot{n}_{\text{CO}_2}$ [mmol/min]	τ [s]	X_C [%]	$\eta_{\text{solar-to-fuel}}$ [%]	U
1	2517	0.63	1119	31.5	17.9	7.1	2.1	52.4	7.9	1.23
2	2499	0.91	1112	33.6	20.2	7.2	1.8	40.2	8.8	1.21
3	2499	1.26	1104	39.6	27.1	7.2	1.5	36.1	10.7	1.21
4	2520	1.57	1099	38.5	28.1	5.9	1.4	28.8	10.5	1.24
5	2752	0.63	1162	38.1	19.4	9.4	2.1	60.7	8.4	1.24
6	2760	0.85	1124	39.1	22.5	8.7	1.9	48.9	9.2	1.22
7	2422	1.25	1094	31.7	22.0	5.5	1.6	29.2	9.2	1.26
8	2412	1.51	1087	38.5	28.7	5.9	1.4	30.5	11.1	1.26
9	2647	0.93	1133	59.9	34.6	13.9	1.7	69.1	13.9	1.26
10	2639	0.94	1142	40.9	21.7	9.8	1.7	44.3	9.2	1.27
11	2641	1.13	1142	60.3	36.9	12.4	1.6	57.8	14.3	1.27
12	2643	1.12	1103	45.8	27.7	10.0	1.7	44.8	10.5	1.27
13	2641	1.25	1129	57.3	35.5	11.9	1.5	50.9	13.8	1.26
14	2633	1.55	1104	48.4	32.2	8.9	1.4	35.3	12.6	1.26
15	2901	0.94	1180	60.3	31.0	15.6	1.7	65.8	11.7	1.25
16	2890	1.14	1175	68.0	35.9	17.4	1.5	61.7	14.4	1.25
17	2874	1.14	1173	60.2	31.8	15.5	1.5	54.7	12.8	1.24
18	2884	1.34	1156	62.1	33.4	16.3	1.4	49.1	14.2	1.24
19	2879	1.36	1145	55.3	31.8	12.6	1.4	43.3	11.5	1.26
20	2864	1.49	1127	56.6	33.2	13.1	1.4	42.0	12.9	1.24
21	3108	1.06	1216	68.5	29.0	20.4	1.5	61.7	11.9	1.25
22	3114	1.35	1173	66.9	30.3	19.2	1.4	48.5	12.6	1.25
23	3112	1.72	1150	86.4	47.4	20.7	1.2	52.5	16.4	1.26
24	3121	2.02	1146	85.7	52.9	17.3	1.1	46.1	16.9	1.27
25	3341	1.42	1194	86.4	43.5	22.7	1.3	61.5	15.5	1.25
26	3328	1.65	1207	83.1	39.6	23.5	1.2	50.5	14.2	1.24
27	3332	1.86	1176	84.0	43.0	21.7	1.1	46.0	14.6	1.25
28	3345	2.03	1153	80.8	44.9	19.2	1.1	41.8	14.8	1.26
29	3566	1.86	1209	87.5	41.0	24.3	1.1	46.4	13.8	1.25
30	3555	2.03	1198	96.1	46.3	25.6	1.0	46.8	15.8	1.26

Figure 3b presents a representative experimental run conducted with the D-I solar reactor. Plotted are the temporal variation of $\dot{m}_{\text{feedstock}}$, $\dot{Q}_{\text{solar}}$, T_{reactor} , T_{shell} , T_{offgas} and the product gas evolution at ambient pressure. The period of the experiment considered to approximate steady-state conditions is indicated by the two vertical dotted lines. For this run, these conditions were: $p = 1$ bar, $\dot{m}_{\text{feedstock}} = 0.94$ g/min, $\text{H}_2\text{O}:\text{C} = 3.24$, and $\dot{Q}_{\text{solar}} = 2.9$ kW, leading to $T_{\text{reactor}} = 1180$ °C, $T_{\text{shell}} = 1061$ °C, $T_{\text{offgas}} = 709$ °C, and yielding a syngas flow of $\dot{n}_{\text{H}_2} = 60.3$ mmol/min, $\dot{n}_{\text{CO}} = 31.0$ mmol/min, and $\dot{n}_{\text{CO}_2} = 15.6$ mmol/min. The performance indicators obtained were: $X_{\text{C}} = 65.8\%$ within $\tau = 1.7$ s, $\eta_{\text{solar-to-fuel}} = 11.7\%$, and $U = 1.25$.

Figure 9a-c present $\dot{n}$ for the species H_2 (■), CO (▲), and CO_2 (●) as a function of T_{reactor} for three slurry feeding rates: a) $\dot{m}_{\text{feedstock}} = 0.91 \pm 0.04$ g/min; b) $\dot{m}_{\text{feedstock}} = 1.10 \pm 0.05$ g/min; and c) $\dot{m}_{\text{feedstock}} = 1.43 \pm 0.07$ g/min. For all runs, $\dot{n}_{\text{syngas}}$ increases with T_{reactor} . As for the quality of the syngas, Figure 9d presents the molar ratios $\text{H}_2:\text{CO}$ and $\text{CO}_2:\text{CO}$ as a function of T_{reactor} for all experimental runs. Both molar ratios increase linearly with T_{reactor} , independent from $\dot{m}_{\text{feedstock}}$, presumably because of the water-gas shift reaction at the exit of the solar reactor. CH_4 , when detected, never exceeded 0.38 mmol/min.

Figure 9 - a-c) Molar evolution for the species H_2 (■), CO (▲), and CO_2 (●) as a function of T_{reactor} for three slurry feeding rates. d) Molar ratios $H_2:CO$ (▼) and $CO_2:CO$ (◆) as function of T_{reactor} , obtained with the D-I solar reactor.

3.3 Comparison of the solar reactor configurations

Figure 10 presents X_C (a) and $\eta_{\text{solar-to-fuel}}$ (b) as function of T_{reactor} for: the I-I solar reactor (●) operated at $p = 1.0$ bar, $\dot{m}_{Ar} = 4$ L_N/min, $\dot{m}_{\text{feedstock}} = 0.82 \pm 0.02$ g/min, and $H_2O:C = 1.73$; and the D-I solar reactor (■) operated at $p = 1$ bar, $\dot{m}_{Ar} = 3$ L_N/min, $\dot{m}_{\text{feedstock}} = 0.91 \pm 0.04$ g/min, and $H_2O:C = 3.24$. Because of the significantly different heat transfer mechanisms that are inherent of the direct or indirect irradiation, different $\dot{Q}_{\text{solar}}$ and $\dot{m}_{\text{feedstock}}$ were required to reach the same T_{reactor} in both solar reactor configurations. Specifically, 11% higher $\dot{m}_{\text{feedstock}}$

was required in the D-I solar reactor to obtain an optically thick medium for volumetric radiative absorption [20], and consequently a higher $\dot{Q}_{\text{solar}}$ was required to heat $\dot{m}_{\text{feedstock}}$ to the desired T_{reactor} . Note also that T_{reactor} is an average temperature measured inside the reaction chamber, but the actual particle temperature, which is pertinent for reaction rates and extents, is unknown. Thus, the comparison in terms of X_C becomes difficult because higher values were obtained for the I-I solar reactor at $T_{\text{reactor}} = 1120$ °C but the contrary is true at $T_{\text{reactor}} = 1200$ °C. X_C increases with T_{reactor} for both reactors, but it is more clearly observed for the I-I configuration because of superior stability of the vortex flow. In contrast, $\eta_{\text{solar-to-fuel}}$ remains constant at around $14.3 \pm 1.1\%$ for the I-I while varied within the range $8.8 - 11.7\%$ for the D-I. Furthermore, despite the less efficient heat transfer mechanism to the reaction site, the I-I solar reactor achieves higher values of $\eta_{\text{solar-to-fuel}}$ over the entire temperature range shown in Figure 10, mainly because the required $\dot{Q}_{\text{solar}}$ to reach the desired T_{reactor} was lower due to the lower $\dot{m}_{\text{feedstock}}$, lower $\text{H}_2\text{O}:\text{C}$ molar ratio, and, as it will be shown in the energy analysis that follows, the lower re-radiation losses. On average, $U = 1.20 \pm 0.04$ and 1.25 ± 0.05 for the I-I and D-I configurations, respectively, and independent of $\dot{m}_{\text{feedstock}}$, $\text{H}_2\text{O}:\text{C}$ molar ratio, or T_{reactor} . As for the syngas quality, the ranges of the $\text{H}_2:\text{CO}$ molar ratio obtained are $1.16 - 1.90$ and $1.34 - 2.36$ for the I-I and D-I configurations, respectively, mainly as a consequence of the $\text{H}_2\text{O}:\text{C}$ molar ratio fed.

Figure 10 - a) Reaction extent, and b) solar-to-fuel energy conversion efficiency as a function of T_{reactor} for the I-I (●) and D-I (■) solar reactors. Experimental conditions: (●) $p = 1$ bar, $\dot{m}_{\text{Ar}} = 4$ L_N/min, $\dot{m}_{\text{feedstock}} = 0.82 \pm 0.02$ g/min, and $\text{H}_2\text{O}:\text{C} = 1.73$; and (■) $p = 1$ bar, $\dot{m}_{\text{Ar}} = 3$ L_N/min, $\dot{m}_{\text{feedstock}} = 0.91 \pm 0.04$ g/min, and $\text{H}_2\text{O}:\text{C}$ molar ratio = 3.24.

Figure 11 presents the energy partition for the I-I and the D-I solar reactors, given as percentage of $\dot{Q}_{\text{solar}}$ averaged over all experimental runs at ambient pressure under approximately steady-state conditions. For the D-I solar reactor, the conductive heat losses are reported as $\dot{Q}_{\text{convection,shell}} + \dot{Q}_{\text{radiation,shell}}$ [20]. $\dot{Q}_{\text{sensible}}$ is higher for the D-I solar reactor as a result of the higher $\dot{m}_{\text{feedstock}}$, $\text{H}_2\text{O}:\text{C}$ molar ratio, and $\dot{m}_{\text{Ar}}$ needed for protecting the window. $\dot{Q}_{\text{conduction,front}}$ is significantly lower in the I-I solar reactor because the water-cooling

refrigeration of the window's mount for the D-I solar reactor was replaced with air-cooling of the windowless front. Further, the higher apparent absorptivity of the I-I solar reactor led to lower $\dot{Q}_{\text{reradiation}}$ at the expense of the additional $\dot{Q}_{\text{convection,cavity}}$. $\dot{Q}_{\text{chemical}}$ is comparable, but as seen in Figure 10 and discussed in the previous paragraph, $\eta_{\text{solar-to-fuel}}$ is higher for the I-I configuration over the temperature range 1000 – 1300 °C.

Figure 11 - Energy partition for the I-I and D-I solar reactors, given as percentage of the solar radiative power input averaged for all experimental runs at ambient pressure and under approximate steady-state conditions.

4. Summary & Conclusions

We experimentally demonstrated for the first time the solar-driven gasification of carbonaceous feedstock at high pressures using a robust design of an indirectly-irradiated solar thermochemical reactor. A peak solar-to-fuel energy conversion efficiency of 20% and almost complete conversion in less than 5 seconds point out to the technical feasibility of the solar reactor technology. For all 51 solar runs, the calorific value of the feedstock was upgraded in the range 16 – 35%, underpinning the benefit of solar-upgrading, in contrast to

the inherent downgrading that results from autothermal gasification. High-quality pressurized syngas with no tars was produced, facilitating its integration to gas-to-liquid processing. Ar was substituted by CO₂, increasing the partial pressure of the gasifying agent and eliminating the consumption of expensive inert gas. Higher reaction extents and comparable solar-to-fuel energy conversion efficiencies and calorific upgrade factors were obtained on average with the indirectly-irradiated solar reactor than with the directly-irradiated solar reactor.

Nomenclature

Latin symbols

C	solar concentration ratio [kW/m ²]
d	particle size [μm]
LHV	lower heating value [MJ/kg]
$\dot{m}$	mass flow rate [g/min], [L _N /min]
$\dot{n}$	molar flow rate [mmol/min]
p	pressure [bar]
$\dot{Q}$	power [W]
R	universal gas constant: 8.314 J/(K mol)
r	radial coordinate [mm]
T	temperature [°C], [K]
t	time [min]
U	energetic upgrade factor
u	gas velocity [m/s]
V	volume [m ³]
X_C	reaction extent [%]
x	H:C molar ratio

y	O:C molar ratio
z	horizontal coordinate [mm]

Greek symbols

$\eta_{\text{solar-to-fuel}}$	solar-to-fuel energy conversion efficiency [%]
τ	nominal residence time [s]
φ	angular coordinate [°]

Subscripts

cavity	cavity
chemical	chemical
conduction	heat conduction
convection	free convection
feedstock	feedstock, charcoal
gi	gradually increasing
i	inlet
N	normal conditions: 273.15 K and 1 atm
offgas	off gas stream
radiation	radiation
reactor	reactor
reflection	reflection
reradiation	re-radiation
sensible	sensible heat
shell	metallic shell
solar	solar
solar-to-fuel	solar-to-fuel

syngas	syngas
v10,50,90	upper threshold particle size in μm

Abbreviations

CCD	charged coupled device
D-I	directly-irradiated
FT	Fischer-Tropsch
IR	infrared
GC	gas chromatograph
HFSS	high-flux solar simulator
I-I	indirectly-irradiated

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