



Delft University of Technology

Microwave-driven plasma gasification for biomass waste treatment at miniature scale

Sturm, Guido S J; Navarrete Muñoz, Alex; Purushothaman Vellayani, A.; Stefanidis, Georgios D.

DOI

[10.1109/TPS.2016.2533363](https://doi.org/10.1109/TPS.2016.2533363)

Publication date

2016

Document Version

Accepted author manuscript

Published in

IEEE Transactions on Plasma Science

Citation (APA)

Sturm, G. S. J., Navarrete Muñoz, A., Purushothaman Vellayani, A., & Stefanidis, G. D. (2016). Microwave-driven plasma gasification for biomass waste treatment at miniature scale. *IEEE Transactions on Plasma Science*, 44(4), 670-678. Article 7438874. <https://doi.org/10.1109/TPS.2016.2533363>

Important note

To cite this publication, please use the final published version (if applicable).

Please check the document version above.

Copyright

Other than for strictly personal use, it is not permitted to download, forward or distribute the text or part of it, without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license such as Creative Commons.

Takedown policy

Please contact us and provide details if you believe this document breaches copyrights.

We will remove access to the work immediately and investigate your claim.

Microwave-driven plasma gasification for biomass waste treatment at miniature scale

Guido S.J. Sturm^a, Alexander Navarrete Muñoz^{a,b}, P.V. Aravind^a, Georgios D. Stefanidis^{c,*}

^aProcess and Energy Department, Delft University of Technology, Leeghwaterstraat 39, 2628 CB Delft, The Netherlands

^bDepartment of Chemical Engineering and Environmental Technology, University of Valladolid, c/Doctor Mergelina s/n, 47011 Valladolid, Spain

^cChemical Engineering Department, Katholieke Universiteit Leuven, Celestijnenlaan 200f, 3001 Leuven, Belgium

* Corresponding author (georgios.stefanidis@cit.kuleuven.be)

Abstract

Gasification technology may combine waste treatment with energy generation. Conventional gasification processes are bulky and inflexible. By using an external energy source, in the form of microwave-generated plasma, equipment size may be reduced and flexibility as regards the feed composition may be increased. This type of gasification may be combined with fuel cell technology to generate electricity for on-site microwave generation. In this work, we present short gasification experiments with cellulose, as model biomass compound, in air plasma. In order to optimize reaction rates, gasification and plasma generation are combined in the same volume in order to expose the solids to plasma of maximum intensity. The heating value of the fuel gas yield exceeds, up to 84%, the net microwave energy transmitted into the reactor over a range of operating conditions. As the system has not been optimized, in particular regarding residence time, the results give confidence that this concept can eventually be developed into a viable small-scale decentralized gasification technology.

Keywords

Microwave plasma, plasma gasification, biomass, cellulose, waste

28 **1. Introduction**

29 In view of the scarcity of energy resources, concern about emissions, and the growing population, two
30 future challenges can be identified. These are the effective exploitation of renewable energy sources,
31 and destruction and/or utilization of organic waste materials. The conversion process that is typically
32 considered for such challenge is gasification, which is a process converting organic feedstock into fuel
33 gas, and which has already been widely demonstrated at large scale.

34 For those situations, however, in which no or very little supporting infrastructure is available, or for
35 cases that require relatively small and mobile installations, no satisfactory solutions do yet exist. This
36 holds for the context of this present study that is supported by the Bill & Melinda Gates Foundation in
37 the context of the “Reinvent the Toilet Challenge” [1]. This challenge aims to provide global
38 sanitation improvement by providing of small and mobile treatment units for human waste materials in
39 developing regions. The requirements also hold for many other conceivable situations that require
40 small scale waste destruction, such as for example local destruction of chemical waste or shipborne
41 waste destruction.

42 Another potential area of application that is gaining attention is small-scale conversion and storage of
43 energy for load leveling of renewable power generation. The IEA Technology Roadmap Energy
44 Storage reports a need for a 310 GW storage capacity to accommodate a 27-44% renewable electricity
45 production in 2050; it also reports that the technologies considered in this context – flywheel
46 technologies, supercapacitors, superconducting magnetic energy storage, battery technology, pumped
47 storage hydropower, and compressed air energy storage – are challenged in terms of high costs, large
48 footprint, and low energy density [2]. Another technology discussed in this context by both the IEA
49 report and Turner [3] are water electrolyzers, hydrogen storage, and fuel cells; as with the other
50 technologies, though, it is pointed out that costs remain a challenge in the foreseeable future. An
51 alternative may be found by enhancing the aforementioned gasification process with renewable
52 energy, thereby storing this energy in the fuel gas that is produced. The specific method of
53 enhancement considered in the context of this study is combining gasification with plasma. A notable
54 aspect of plasma is that in principle it can respond fast to intermittent and fluctuating energy demand
55 and supply. Downscaling will remain an important consideration for this kind of application, though.
56 Typically, process systems loose dynamic responsiveness as they get larger, so exploring the
57 downscaling limitations will remain worthwhile.

58 Gasification is a process in which solid feedstocks such as biomass and coal are made to react with a
59 gasification agent such as air, steam or oxygen under sub-stoichiometric conditions resulting in the
60 formation of syngas, i.e. a mixture of hydrogen and carbon monoxide, which is an easy to handle
61 gaseous fuel and which forms a building block for production of storable fuel and chemicals.
62 Conventionally, gasification is carried out using different reactor types such as fixed bed gasifiers,
63 fluidized bed gasifiers or entrained flow gasifiers [4]. However, these reactor types are used in
64 general to process easily gasified feedstocks such as clean wood, low ash content coal etc. While small
65 scale gasification is achieved using fixed bed reactors for example in cooking applications [5,6], well
66 prepared wood chips or pellets are in general required for ensuring smooth operation of the reactors.
67 Furthermore, conventional gasification requires part of the feedstock to be combusted to provide the
68 process with sufficient thermal energy. The syngas produced can be used for several purposes, it can
69 serve as a raw material for chemicals manufacture, but in the context of this study its utilization in
70 energy generation, conversion and storage is considered. At large scale conventional syngas based
71 thermal power production systems are in principle feasible [7], but at small power levels these are
72 inefficient. It has been shown that SOFCs can operate at small power levels (few kW) with very high
73 efficiency [8]. System studies have also indicated that, biosyngas, once cleaned, can be fed to small-
74 scale SOFCs or SOFC integrated systems so as to produce electric power at high efficiencies [9,10].
75 Further studies have been conducted into the influence of biomass derived contaminants on SOFCs
76 [11,12], gas cleaning systems have been developed [12] and successful gasifier-SOFC integrated
77 operation has been achieved [13]. Present research efforts aim to keep extending the operational
78 flexibility of the gasification process in terms of size and feedstock.

79 **Plasma enhanced gasification**

80 The introduction of thermal plasma into the gasification process may be the key towards process
81 downscaling and enhancing flexibility. The external energy source – the electromagnetic field that
82 energizes the plasma – provides an additional degree of freedom to the process that enables process
83 adaption to feedstock variations in terms of both feeding rate and composition. In case of plasma, the
84 external energy source originates in electrical power, possibly from renewable sources. This avoids the
85 requirement to provide energy to the process by means of partial combustion of the feedstock. In
86 principle, dilution of product gasses with combustion products can thus be avoided; in addition,
87 renewable energy can be stored as heating value of the product gasses. Further, plasma enables intense
88 conditions 4000–5000 K in case of a microwave plasma and even higher at 10000–12000 K in case of

89 an ICP or arc discharge plasma [14]. The intensity allows for rapid processing, lower residence time
90 required, and thus a volumetric process scale-down or miniaturization; furthermore, the intensity
91 enables utilization of feedstocks that are resilient to thermal processing, such as municipal solid waste
92 [15], and do not readily break down in conventional gasifiers. In addition, plasma has been shown to
93 facilitate the reforming – and thereby destruction – of tar that is commonly formed in gasification and
94 that is generally considered problematic [16]. Finally, the additional energy source provides a means
95 to compensate for heat losses that become more constricting as the size of conventional – non-plasma
96 – gasification systems is reduced. Naturally, heat losses compromise efficiency, but below a certain
97 geometrical size threshold – as they become more dominant in the overall energy balance – they might
98 also inhibit conventional gasification processes, because the process will no longer be able to sustain
99 its operational temperature.

100 At large scale, the technical feasibility of plasma gasification is demonstrated by Willis et al. [17]. The
101 authors reported on the design and operation of a large (104 MW fuel gas output) plasma-assisted
102 gasification plant for waste materials. It concerns a system that uses high power (3.22 MWe) arc-
103 plasmas, and thus it demonstrates the general technological and operational feasibility of plasma-
104 assisted waste gasification. Further, a theoretical framework is being developed with several
105 researchers publishing regularly on the subject [18–20]. Other relevant experimental work at smaller
106 scale has been published by Moustakas et al. [21], Lemmens et al. [22], Hong et al. [23], and Uhm et
107 al. [24]. The latter two publications are relevant in the context of this present research since in they
108 report on operation of and gasification with a 915 MHz microwave generated plasma. The latter
109 publication reports on brown coal gasification by microwave-generated steam plasma; a reasonably
110 high cold gas efficiency of 84 % is reported for a medium size gasification unit (500 kW fuel gas
111 output). A number of these authors were also involved in studies on smaller scale 2.45 GHz
112 microwave plasma gasification of brown coal [25] and coal [26] with 8 kW of fuel gas output, and –
113 due to scaling laws – a lower cold gas efficiency at 43 %. Further work at the same scale was done by
114 Yoon and Lee [27, 28], which saw successful gasification of coal and charcoal.

115 In a collaborative effort on the development of the aforementioned project on processing of human
116 waste materials, Liu et al. [29] present a system integration study of a small-scale ~10 kW fuel gas
117 output plasma gasifier integrated with a solid oxide fuel cell (SOFC) stack, a pre-treatment stage for
118 drying and grinding, and a gas cleaning stage to prepare the syngas for the SOFC. It is shown by
119 means of simulation that such a combined system for decentralized treatment of human waste may

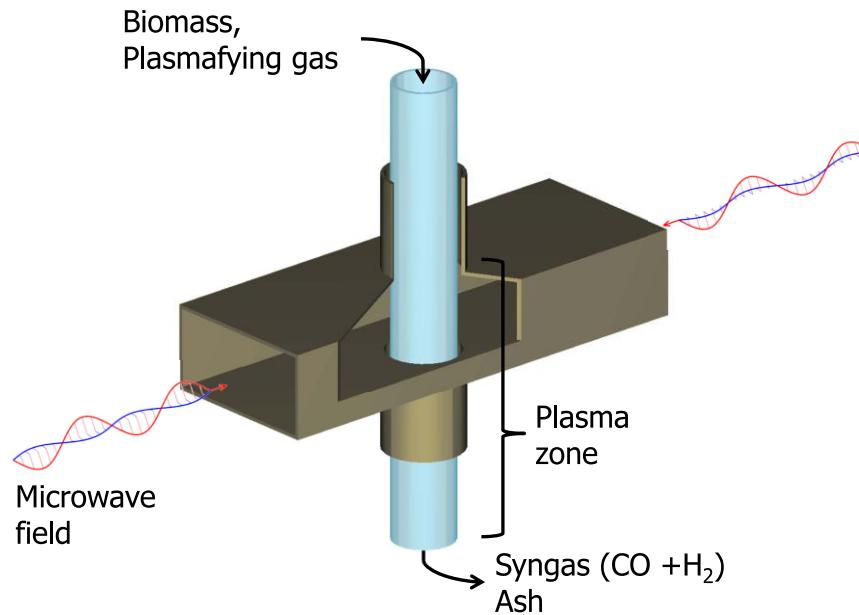
120 indeed form an energetically self-sustained process, which would enable the objective of developing a
121 small-scale processing facility for sanitation waste that is non-reliant on pre-existing infrastructures
122 (i.e. water supply, sewage, electricity supply, and fuels). The feasibility of fuel cell integration was
123 also explored by another group, Galvita et al. [30], and shown to have potential. The purpose of this
124 present study is to focus on the plasma gasification step in the context of the above collaborative effort
125 to develop the larger treatment and energy recovery system.

126 The specific requirements of the intended application pose several tough challenges: the system has to
127 be small, so that heat losses are relatively large and efficiency is limited; the infrastructure limitations
128 do not leave much freedom in terms of plasma agent, as only air is essentially available; and only
129 technologies that in principle are robust and can be manufactured at low cost can be considered.

130 It is not known what the limits are to which the plasma gasification process can be downscaled while
131 retaining adequate effectiveness, but in order to achieve the highest processing rate we expose the
132 feedstock to the plasma at its spatially highest intensity. Specifically, we combine the gasification
133 process and plasma generation in the same part of the reactor, so that gasification occurs in plasma at
134 its maximum intensity.

135 In order to meet the robustness and cost requirements we use a microwave field, which is preferred
136 over an arc discharge plasma or an inductively coupled plasma, because magnetron vacuum tubes are
137 more cost-effective and fit the power required by our application better than the alternatives, while still
138 providing a reliable means to generate an electromagnetic field. In addition, in contrast to arc
139 discharge plasma, microwave generated plasma avoids direct contact of the plasma with electrodes, so
140 electrode wear is avoided.

141 At present, the interplay between the physical phenomena occurring in the plasma gasification reactor
142 thus specified is not known, and design strategies for developing such reactors do not yet exist. Hence
143 we start our exploration with off-the-shelf parts in order to verify the feasibility of the process. Similar
144 to Konno et al. [31] we use cellulose as a model substance for waste materials.

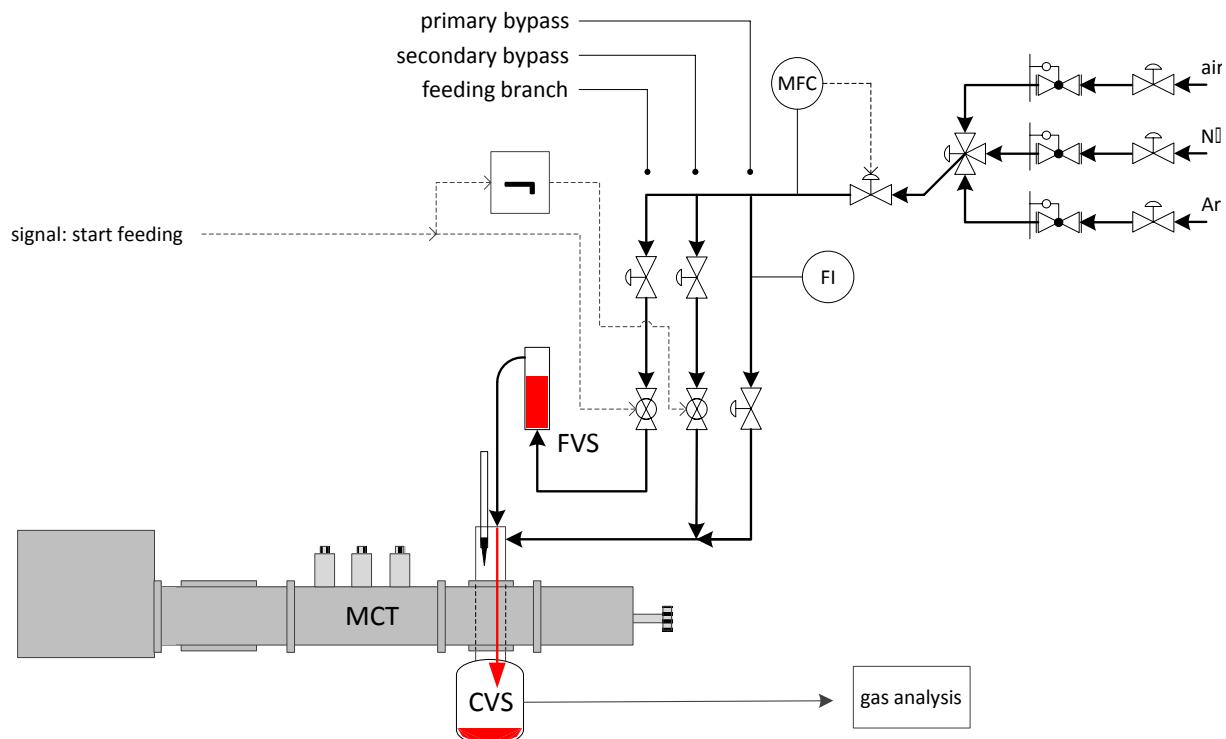


145

146

Figure 1. Schematic of the plasma gasification reactor assembly.

147



148

149
150

Figure 2. Schematic of the experimental system. MCT: microwave circuit; FVS: feeding vessel; CVS collection vessel; MFC: mass flow controller; FI: flow indicator.

151

152

153 **2. Experimental apparatus and methodology**

154 The gasification reactor is essentially constructed out of a quartz pipe with an internal diameter of 31
155 mm and a wall thickness of 2 mm that crosses the broad face of a WR-340 waveguide. Figure 1
156 presents a schematic of the assembly; the quartz pipe is oriented vertically and the waveguide is placed
157 along the horizontal plane. Metal pipes run along the quartz pipe both from the top and the bottom
158 onto the waveguide wall. These pipes are dimensionalized with an internal diameter that is narrow
159 enough to block microwave transmission at the wavelength that the system operates at; thus, the
160 microwave field is contained in the waveguide, while gas and solids are free to flow through the quartz
161 pipe. Further relevant dimensions of the reactor assembly are listed as follows: the cavity formed by
162 waveguide is at the location of the reactor 71 mm high; the section of the reactor passing through a
163 metallic cut-off pipe directly downstream of the cavity is 100 mm long; this is followed by roughly 60
164 mm leading into the collection vessel (CVS, see below). The assembly of plasma generator and reactor
165 are based on, and modified from the commercially available *Downstream Plasma Source* obtained
166 from Sairem SAS [32]. Due to the high temperatures involved (4000-5000 K, [14]) and the presence
167 of a strong microwave field, in situ temperature measurement is not possible in the reactor. The quartz
168 pipe containing the hot plasma is contacted on the outside by air and the waveguide parts at room
169 temperature; a thermal balance between heating and cooling is maintained, so that melting of the pipe
170 is avoided. It did require a significant effort of testing and repairing before suitable operating
171 conditions were found.

172 The reactor operated at ambient pressure; i.e. its outlet is at ambient pressure and due to the flow rates
173 and piping diameters involved, only negligible pressure drop occurs in the system. In the context of
174 this study air is fed from the top into the quartz reactor pipe both as a plasma agent and oxidizer.
175 Before plasma ignition, an ignition electrode system is lowered into the quartz pipe down to the level
176 of the waveguide. Once microwave field generation starts, an initial discharge on the electrode absorbs
177 electromagnetic energy from this field. This discharge then grows into a plasma flame that is blown
178 downwards with the gas stream. After ignition, the electrode is retracted from the reactor and plasma
179 is sustained as long as the microwave field is present.

180 Once plasma has been ignited, solid biomass is also fed from the top into the quartz pipe. As it passes
181 through the plasma flame, the high temperatures and reactive plasma species cause the biomass to

182 gasify, which results in the production of fuel gas in the form of hydrogen, carbon monoxide and
183 methane.

184 Figure 2 presents a schematic of the experimental system. The microwave part of the system is an
185 electromagnetic circuit in typical layout [33] that is constructed out of several WR-340 waveguide
186 elements. The microwave system operates at a frequency of 2.45 GHz, which is compatible with the
187 WR-340 waveguide standard. This frequency was chosen due to the relative compactness of the
188 waveguide elements. The reactor assembly is an integral part of this microwave circuit (MCT) that in
189 all consists of the following parts:

- 190 - A 2.45 GHz microwave generator with a maximum output power of 6 kW.
191 - An isolator, which is a microwave circuit element that protects the microwave generator from
192 exposure to a reflected microwave field. More specifically, it transmits a microwave field
193 traveling in the forward direction, but absorbs it when it travels in the reverse direction.
194 - An impedance transformer, which is a circuit element that can be used to tune the microwave
195 field in the circuit such that no reflections occur towards the generator. Essentially, proper
196 tuning causes the energy of the microwave field to be largely dissipated by energizing the
197 plasma, thus enabling good utilization efficiency of the microwave energy.
198 - The reactor assembly, as described above.
199 - A variable reflector that can be used to position the standing microwave field in the
200 microwave circuit. Tuning of this element allows the positioning of a microwave field
201 maximum at the plasma flame, facilitating energy transfer from the microwave field to the
202 plasma.

203 All microwave circuit parts were obtained from Sairem SAS [32]. This includes the waveguide parts
204 of the reactor assembly, which were later modified to better suit our needs. Figure 3 shows a
205 photograph of the setup layout.

206 The flow system is configured as follows:

- 207 - Gas enters the system through a mass flow controller (MFC, Bronkhorst F-201AV-50K) with
208 which the total gas flow is regulated.
- 209 - After the MFC, the flow is split into three branch lines. Each branch includes a needle valve
210 that enables adjustment of the flow rate through it. One branch line flows to the feeding vessel
211 (FVS, discussed below) while the two other branches bypass the feeding vessel and flow
212 directly into the reactor.
- 213 - Of the bypass branch lines, the primary one includes a flow indicator (FI). Both the secondary
214 bypass line and the branch line that flows to the feeding vessel include an automatic valve.
215 These automatic valves are controlled such that if one opens, the other one closes. The needle
216 valves in the separate branch lines are balanced such that the total flow rate through the MFC
217 is not affected once the automatic valves switch the gas flow from one branch line to the other.
218 The flow indicator can be used to verify this.
- 219 - Initially, the feeding vessel is loaded with solid biomass powder and the branch line to the
220 feeding vessel is closed so no gas flows through the feeding vessel. The secondary bypass line
221 is opened. Once a signal is generated to start feeding, the branch line to the feeding vessel
222 opens and the secondary bypass line closes. The gas flow enters the feeding vessel in the
223 bottom side; the upward flow through the powder bed fluidizes this bed and gradually carries
224 the entrained particle bed into the reactor.
- 225 - After the particles pass the plasma zone in the reactor, the remains are captured in a collection
226 vessel (CVS). The gas flow exits the reactor and passes a sample point before being disposed
227 in ventilation. The sample point has a multi-way valve with three ports to mount gas sample
228 bags on.

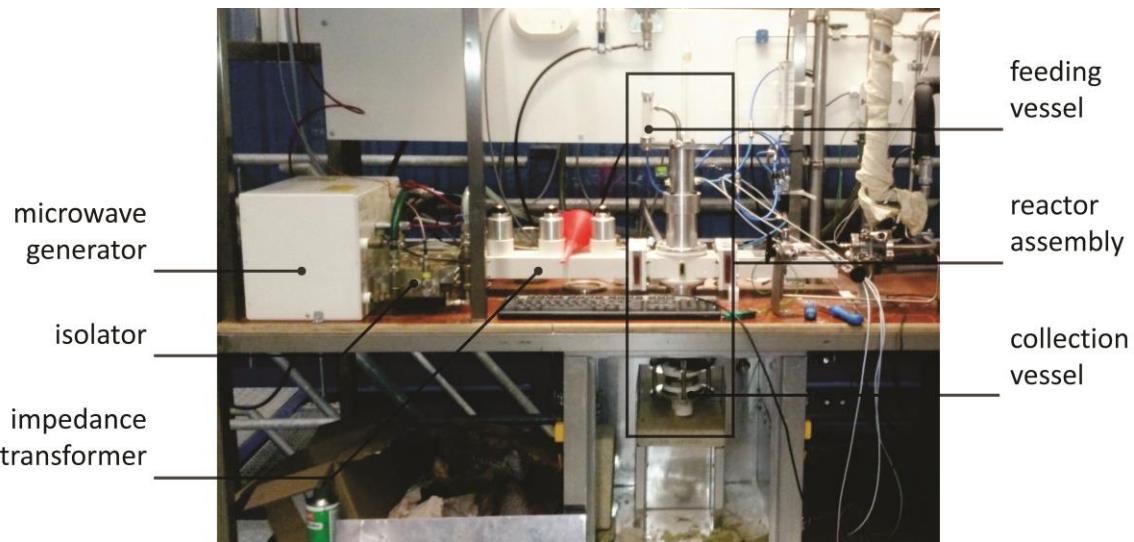
229 In all, the experimental procedure is as follows:

- 230 - Preparatory experiments are conducted to adjust the needle valves to the desired settings, i.e.
231 they are set such that 1) the total flow rate is little affected by switching from the secondary
232 bypass line to the feeding vessel line, and 2) the desired solids flow rate is achieved once this
233 switch has occurred.
- 234 - The feeding vessel is loaded with a weighed amount of powdered solid biomass. The gas line
235 to the feeding vessel is closed and the secondary bypass line is opened.

- 236 - The total flow rate is set with the MFC; a set point value for microwave generation is given to
237 the microwave generator though microwave generation does not start at this point.
- 238 - The ignition electrode is lowered to the level of the microwave zone. Then, microwave
239 generation is started and after plasma ignition, the ignition electrode is retracted.
- 240 - Forward and reflected power are automatically monitored and logged. Once the plasma
241 ignition signature in these signals is detected by the control system, solids feeding starts
242 automatically after a timed interval by switching gas flow from the secondary bypass line to
243 the feeding vessel line.
- 244 - After the experiment, the remaining solid biomass powder in the feeding vessel is collected
245 and weighed. Dividing the mass difference – the mass fed to the reactor – by the total process
246 time yields the feeding rate of the solids.
- 247 - The gas samples collected in the bags are analyzed by gas chromatography (Varian CP4900).

248 All automated procedures were implemented in a LabView 2010 [34] environment.

249

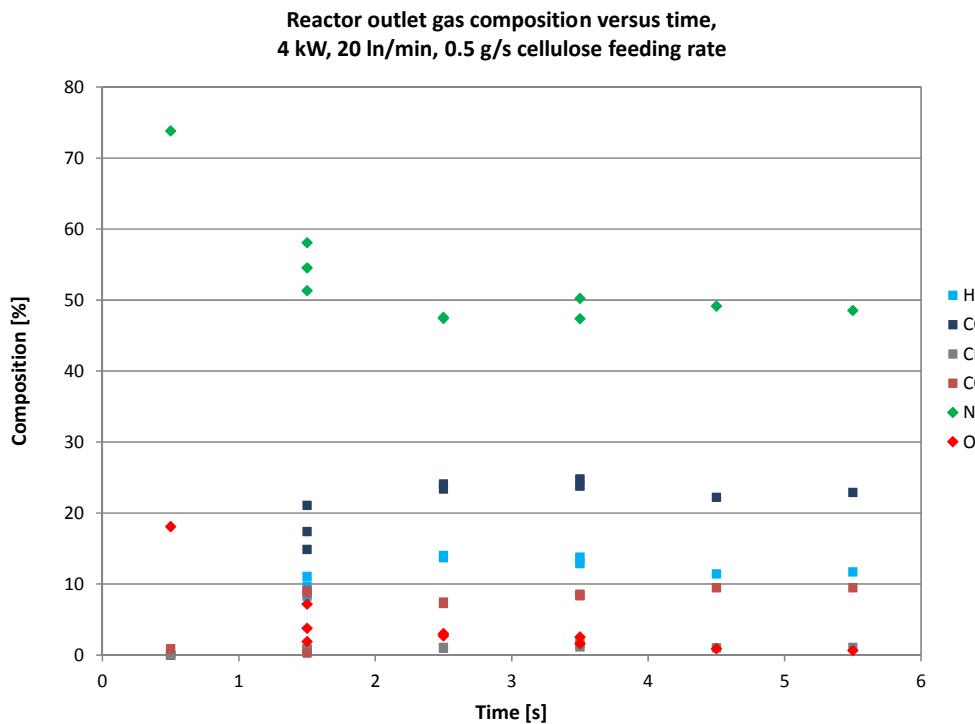


250

251

252

Figure 3. Photograph of setup with main parts indicated.

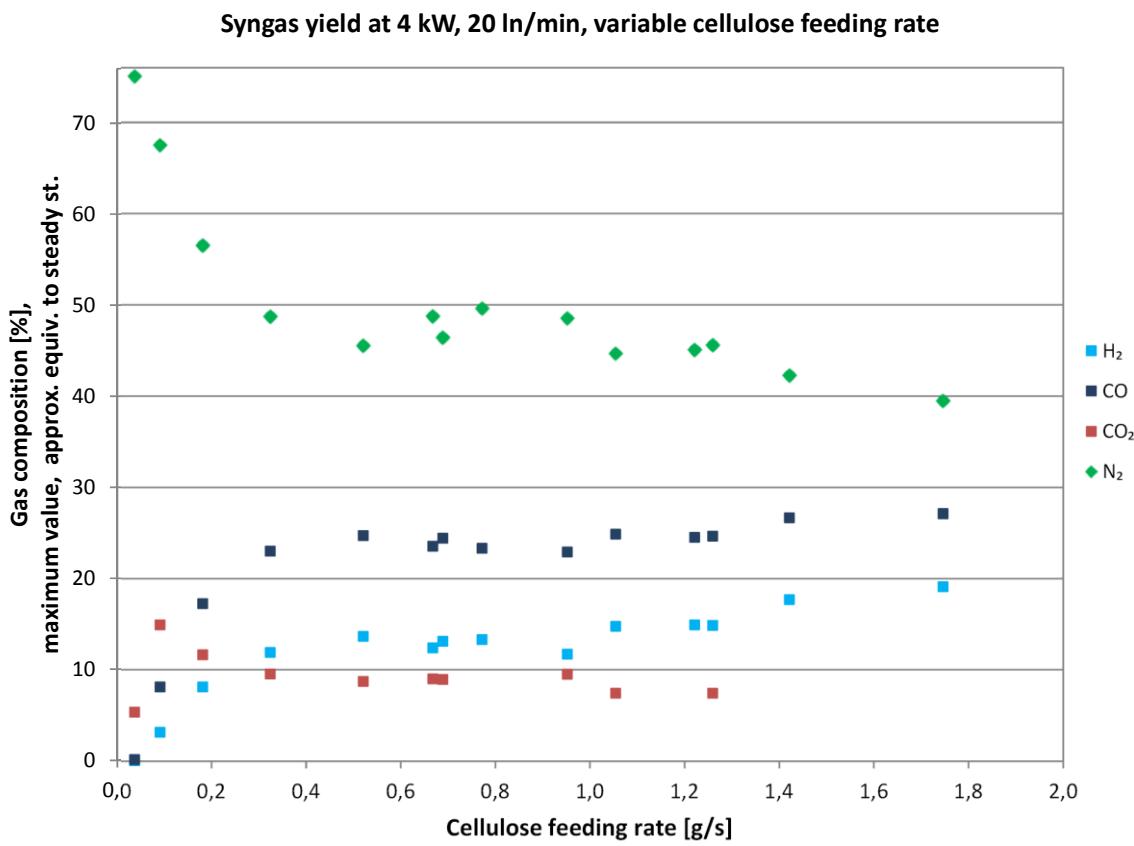


253

254

255

Figure 4. Transient product gas composition (mol%).



256

257 Figure 5. Quasi-steady state fuel gas composition (mol%) at the reactor outlet vs. cellulose feeding rate.

258

259 The experiments were conducted with air as a plasma agent and cellulose ($(C_6H_{10}O_5)_n$, obtained from
260 Aldrich, microcrystalline, 10-350 μm measured with Microtrac S3500) as a model biomass compound.
261 During preparatory experimentation it was found that the process appeared to perform best around the
262 following parameter settings: a feeding rate of air of 20 natural liters (i.e. at 0 °C and 1 atm) per
263 minute (ln/min); a forward microwave power of 4 kW; and a cellulose feeding rate of 0.5 grams per
264 second; this amounts to an air to fuel equivalence ratio of 0.17. These conditions were the starting
265 point for a subsequent parametric study. Repeated experiments were conducted with the same process
266 parameters in terms of microwave power, air feeding rate and cellulose feeding rate, but with varied
267 timing of 1 sec gas sampling intervals so as to obtain transient process data. Figure 4 presents the
268 transient reactor outlet gas composition, i.e. the gas compositions of the samples versus the respective
269 time intervals.

270 In order to approximate steady state continuous operation, it is assumed that the peak concentration of
271 fuel gas in the gas composition would correspond to the steady state value. For the graph in Figure 4,
272 this would correspond to a yield of 14 mol% H_2 and 25 mol% CO and 1.5 mol% CH_4 at a microwave
273 power of 4 kW, an air feeding rate of 20 ln/min and a cellulose feeding rate of 0.5 g/s. The flow rate of
274 producer gas at the outlet is calculated by multiplying the inlet air flow rate with the ratio of the (inert)
275 nitrogen concentration at the inlet to its concentration at the outlet; in Figure 4 this outlet flow rate
276 would correspond to 32 ln/min. By varying the cellulose feeding rate, the relation of gas composition
277 versus cellulose feeding rate is obtained and shown in Figure 5; here the cellulose feeding rate varies
278 from 0.05 to 1.75 g/s, while air feeding rate and microwave power remain constant at 20 ln/min and 4
279 kW.

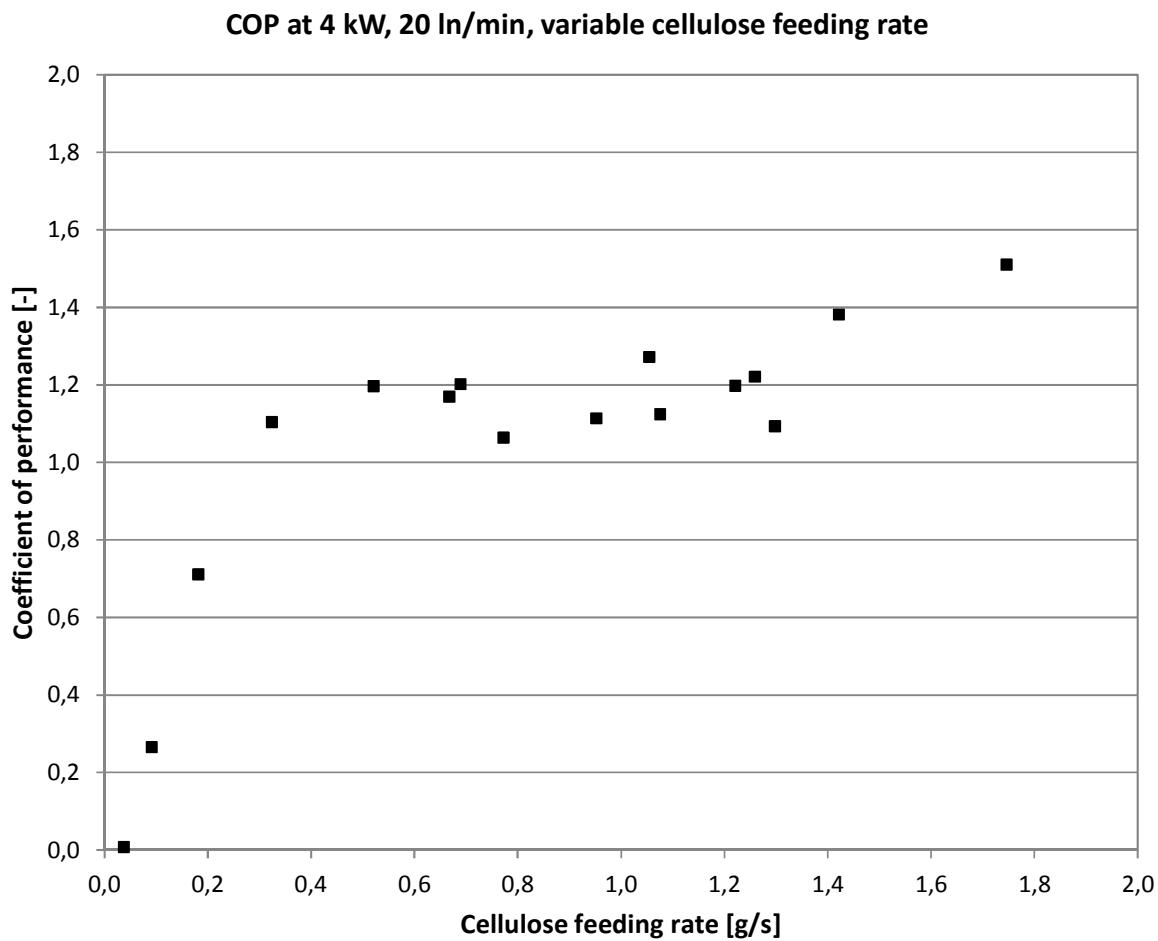
280

281 **3. Results and discussion**

282 From the aforementioned plots, a number of process characteristics can be derived. A general feature,
283 notable in both Figure 4 and 5, is the relatively clear trends that the data points form. This indicates
284 that for these particular experimental conditions, the process is reproducible and stable. In addition,
285 the process is dynamically fast, which is apparent from the graphs of the gas composition transients in
286 Figure 4, as they reach the maximum fuel gas yield in only a few seconds. Further, methane
287 concentration is relatively low (~1.5 mol%). Finally, as mentioned above, the yield of producer gas is
288 roughly 30 ln/min.

289 The most notable feature in the trend of the fuel gas concentration versus cellulose feeding rate (Figure
290 5) is the initial fast rise followed by stagnation for cellulose feeding rates beyond 0.5 g/s. It appears
291 that this plateau-like trend in the fuel gas yield occurs because of the short residence time that is
292 estimated at 20 to 50 ms. More specifically, as the solids feeding rate increases, the fuel gas
293 production may rise, but this will also increase the gas volume and flow velocity. This would reduce
294 the residence time and the process effectiveness. It is further noted here that raising the microwave
295 power does not improve performance; although more power would increase temperature and thus the
296 rate of heat and mass transfer and reaction rates, gas expansion would also accelerate the flow, again
297 cutting short the residence time.

298 The incomplete conversion is also apparent from the cellulose collected in the collection vessel. As
299 cellulose contains no inorganics, full conversion into gas is possible provided sufficient contact time
300 with plasma. In contrast though, the material collected in the collection vessel still resembles cellulose,
301 albeit with a light yellow/brown hue suggesting light tar deposition. Due to the nature of the
302 experimental procedure, a precise mass balance over the reactor could not be established. In particular,
303 transient effects hinder calculation of an accurate figure based on the carbon balance between the
304 cellulose that was fed and the outlet gas yield; moreover, the mass balance over the feeding vessel and
305 collection vessel is considered imprecise, because part of the solids may have been carried further
306 downstream by the gas flow. Nevertheless, from the available experimental data presented in Figure 5,
307 an estimate for the conversion was made based on the carbon balance. It was found that at a cellulose
308 feeding rate of 0.5 g/s, approx. 45 % of the cellulose mass is converted into fuel gas, which
309 corresponds to 0.24 g/s in absolute terms. Further, at 1.7 g/s feeding rate the conversion has dropped to
310 15 %, corresponding to 0.26 g/s conversion rate. Overall, between the 0.5 and 1.7 g/s feeding rates, the
311 conversion rate appears steady between 0.21 and 0.26 g/s and in agreement with the plateau-like trend
312 described above. The exact cause of the limited conversion cannot be stated conclusively. Because of
313 the specific interactions between plasma flame, microwave field, and gas flow patterns, the plasma
314 does not distribute evenly over the cross-section of the quartz tube, which could mean that cellulose
315 partially bypasses the plasma flame. However, heat and mass flow limitations between the plasma and
316 the individual cellulose particles might also occur due to the short contact time (20-50 ms) and the low
317 density of plasma.



318

319 Figure 6. Coefficient of performance versus cellulose feeding rate at forward microwave power of 4 kW and air
320 feeding rate of 20 ln/min.

321

322 Enhancing the residence time by increasing the process volume would appear to be a straightforward
323 way to improve conversion and, ultimately, waste destruction. The reactor presented in this study is a
324 modified commercial off-the-shelf device that is not optimized for this gasification process. As a first
325 design iteration, it demonstrates the feasibility of plasma gasification at this scale, though it is
326 decidedly non-optimal. Subsequent design iterations will improve performance further, for example by
327 applying an electromagnetic field with a longer wavelength or combining multiple plasma generators
328 in series in the gasification system.

329 Despite the issues related to conversion and residence time, the chemical energy produced by the
330 process gives a promising outlook in the context of the intended application – small- or miniature-
331 gasification for decentralized waste treatment. Figure 6 presents the coefficient of performance of the
332 gasification process for the experimental conditions and outlet gas compositions in Figure 5. In this
333 context, this coefficient is defined as the ratio of the chemical energy contained in the produced fuel
334 gas versus the net microwave transmission as described by the following formula:

$$\text{coefficient of performance} = \frac{\frac{Q_{in}}{V_m} \frac{x_{N_2,air}}{x_{N_2,out}} \left(x_{H_2,out} \Delta H_{LHV,H_2} + x_{CO,out} \Delta H_{LHV,CO} + x_{CH_4,out} \Delta H_{LHV,CH_4} \right)}{P_F - P_R} \quad (1)$$

335 Here Q_{in} is the volumetric flow rate of air into the reactor; V_m is the gaseous molar volume; $x_{N_2,air}$ is the
336 mole fraction of nitrogen in air; $x_{N_2,out}$, $x_{H_2,out}$, $x_{CO,out}$ and $x_{CH_4,out}$ are the mole fractions in the reactor
337 outlet stream of N₂, H₂, CO and CH₄ respectively; $\Delta H_{LHV,H_2}$, $\Delta H_{LHV,CO}$ and $\Delta H_{LHV,CH_4}$ are the molar
338 lower heating values of H₂, CO and CH₄ respectively [35]; P_F is the forward microwave power and P_R
339 is the reflected microwave power. The molar fuel gas output is calculated relative to the nitrogen flow;
340 the fuel gas flows are then multiplied with the respective lower heating values; the sum of these
341 products is then divided by the difference of the forward microwave power and the reflected
342 microwave power. This is the aforementioned ratio of chemical energy in the fuel gas versus net
343 microwave transmission towards the plasma reactor. Table 1 presents the values of the parameters in
344 Eq. 1 for the outlet gas compositions in Figure 5 at a solids feeding rate of 0.52 g/s.

346

Table 1. Parameters in the calculation of the coefficient of performance, for the gas composition presented in Figure 5 at a solids feeding rate of 0.52 g/s.

Q_{in}	20	ln/min
	0.33	l/s (0 °C, 1 atm)
V_m	22.41	l/mol (0 °C, 1 atm)
$x_{N_2,air}$	78.08	%
$x_{N_2,out}$	45.52	%
$x_{H_2,out}$	13.65	%
$x_{CO,out}$	24.7	%
$x_{CH_4,out}$	1.15	%
$\Delta H_{LHV,H_2}$	240.8	kJ/mol [35]
$\Delta H_{LHV,CO}$	282.6	kJ/mol [35]
$\Delta H_{LHV,CH_4}$	801.8	kJ/mol [35]
P_F	4	kW
P_R	1.61	kW
<i>COP</i>	1.19	-

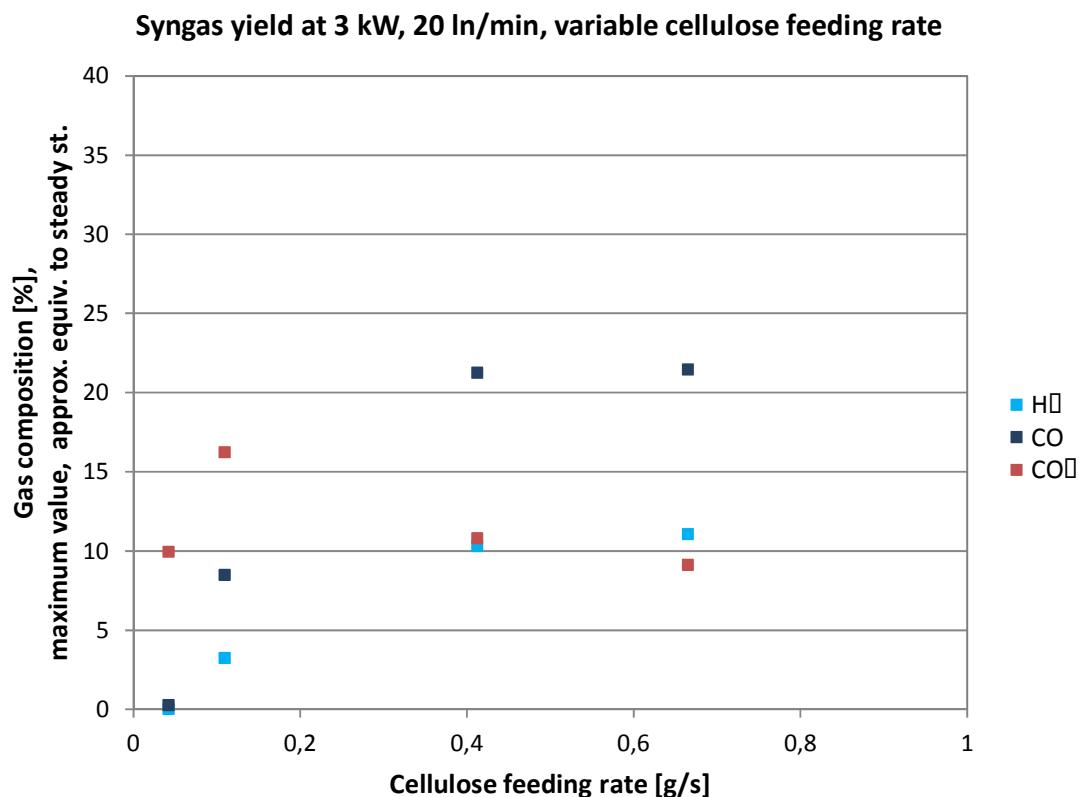
349 It can be seen in Figure 6 that the coefficient of performance exceeds unity for cellulose feeding rates
 350 greater than ~0.3 g/s, while it fluctuates around 1.2 for feeding rates between ~0.3 and ~1.5 g/s.
 351 Beyond 1.5 g/s, the coefficient appears to be rising further to ~1.5. These results are promising
 352 because although the process is non-optimized, there already is a surplus of energy. We expect that
 353 process improvement, most notably on the residence time aspect, will enable better conversion, higher
 354 release of fuel gas, and enhanced energy surplus. The methods noted above could be a starting point
 355 towards this objective. Further, improved management of radiative heat losses could be applied to
 356 improve process performance.

357 The process was further investigated by varying the microwave power and air flow rate parameters in
 358 relation to the base case described above. The procedures above were repeated with A) the microwave
 359 power reduced to 3 kW and the same flow rate of 20 ln/min, B) proportional reduction in both
 360 parameters (i.e., microwave power of 3 kW and flow rate of 15 ln/min) and C) reduction in only the
 361 flow rate to 15 ln/min, while retaining a 4 kW microwave power. The resulting graphs of gas
 362 composition vs. cellulose feeding rate are presented in Figures 7a to c, respectively.

363 The graphs at reduced power (3 kW, Figures 7a-b) show similar trends as Figure 5, with the one in
364 Figure 7a being slightly lower. This indicates that the processes are comparable. One major difference
365 is in the stability of the plasma flame. In the cases of Figures 7a and 7b, the plasma would quench, i.e.,
366 the lower intensity microwave field would not sustain it, much more often than in the case presented in
367 Figure 5. This instability is also apparent from the fact that the highest cellulose flow rate at which
368 plasma could be sustained in Figures 7a-b is ~0.7 g/s, while in Figure 5, it is ~1.8 g/s. A mechanism
369 that may be at play here is that the endothermicity of the process cools down the plasma, which
370 reduces the electron density and electrical conductivity of plasma. Consequently, less microwave
371 energy would be absorbed, resulting in progressive reduction of temperature up to the point where
372 ionization no longer takes place and plasma generation stops.

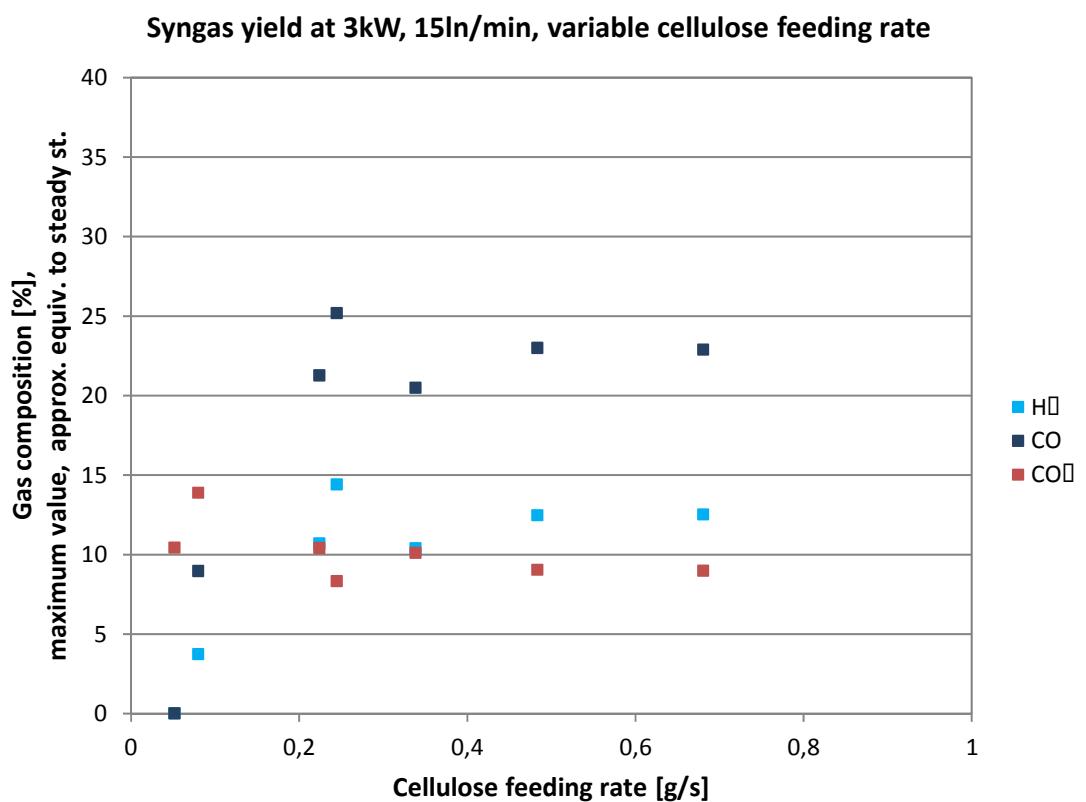
373 In Figure 7c, this type of instability is also present – no plasma can be sustained beyond roughly 0.87
374 g/s –, though another type of instability is much more apparent. In Figures 5 and 7a-b, the trends
375 formed by the data points are fairly clear, whereas in Figure 7c, a much larger spread in the gas
376 composition can be observed. Strikingly, within this set of experiments, at the same experimental
377 conditions, the best yield of 35 % CO and 24 % H₂ is contrasted to the worst yield of 17 % CO and 10
378 % H₂. The latter experiment being an actual attempt to repeat the former with the same settings in
379 terms of microwave power, air flow rate and valve settings. The feeding rate of cellulose in both cases
380 is around 0.4 g/s, though not exactly the same due to the limitations of the solids feeding method
381 employed.

382



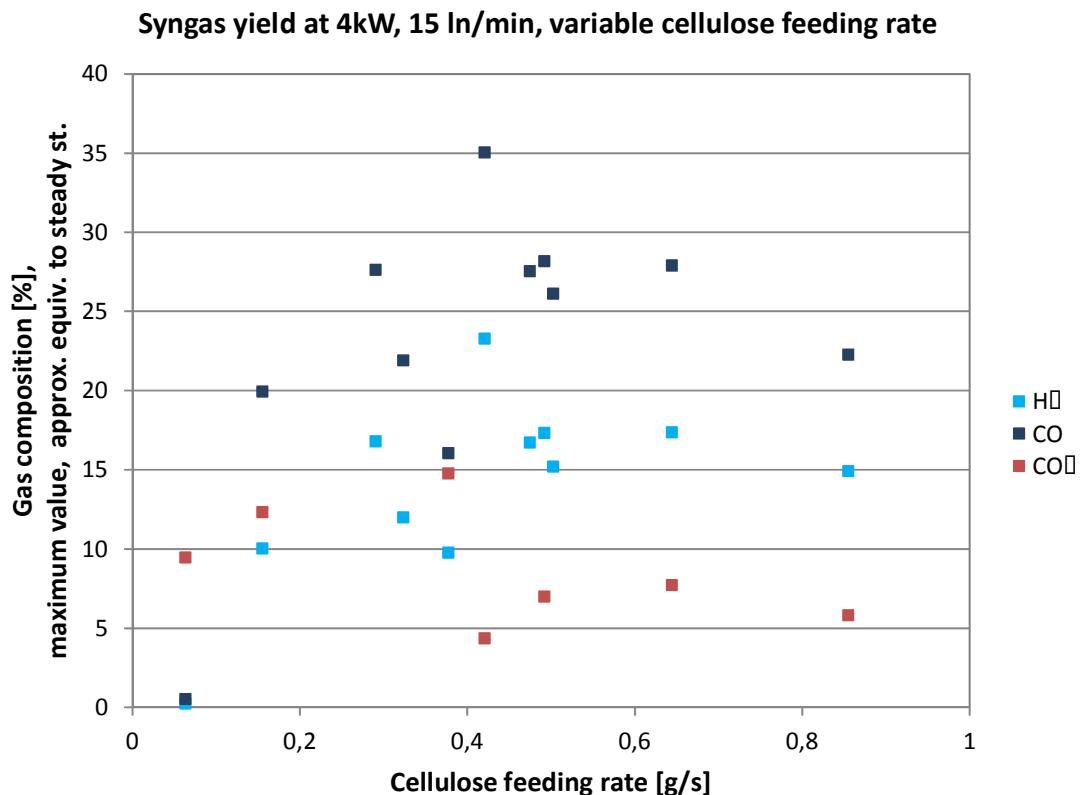
383

(a)



384

(b)



385

(c)

386 Figure 7. Quasi-steady state fuel gas composition (mol%) at the reactor outlet at vs. cellulose feeding rate under
 387 variation of forward microwave power and air flow rate: a) 3 kW and 20 ln/min; b) 3 kW and 15 ln/min;
 388 c) 4 kW and 15 ln/min.

389

390

391 Table 2. Maximum coefficient of performance from the data
 in Figures 5 and 7a-c in the 0-1 g/s cellulose feeding rate
 interval.

Microwave power	Air flow rate	Coefficient of performance
3 kW	15 ln/min	1.19
3 kW	20 ln/min	1.30
4 kW	15 ln/min	1.84
4 kW	20 ln/min	1.20

392

393 Table 2 presents the maximum coefficients of performance in the 0 to 1 g/s interval of the cellulose
 394 feeding rate for the variations in microwave power and gas flow rate presented in this work (Figures 5,
 395 7a-c). For the cases of 4 kW and 20 ln/min; 3 kW and 20 ln/min; and 3 kW and 15 ln/min, the
 396 coefficients of performance are comparable at ~1.2–1.3. For the case of 4 kW and 15 ln/min, the
 397 maximum coefficient of performance is 1.84; i.e. *84% more energy* is contained in the heating value of
 398 the fuel gas obtained than was applied by the net microwave energy input. Despite this being a single
 399 case, it does further strengthen the confidence that the gasification process may attain an energy
 400 recovery sufficiently high to allow for energetically self-sufficient integration with a gas cleaning/fuel
 401 cell system downstream. The instabilities and fluctuations observed also point out the need to gain an
 402 intricate understanding of the complex interrelating phenomena that occur in this gasification process.
 403 Design and optimization of this reactor requires insight into the microwave field/plasma interactions,
 404 the dynamics of gaseous and powder flow, the heat transfer and chemical conversions, as well as into
 405 the manner in which these aspects interrelate.

406

407 **4. Conclusions**

408 In this study we evaluate a 2.45 GHz microwave plasma system for single pass gasification of solid
 409 biomass. The application context for this system is small-scale waste destruction, energy recovery, and
 410 renewable energy storage in the form of chemicals. We combine gasification and plasma generation in
 411 a single volume to maximize process intensity and speed, and to minimize equipment size. This nature
 412 of the work is exploratory to investigate process feasibility. Since it uses a modified off-the-shelf
 413 plasma generator, not a design optimized for this process, there is room to improve performance.
 414 Notably, it was found that the residence time in the system is too short for full biomass conversion.
 415 Design approaches to improve this have been suggested. We found that plasma stability poses a

416 challenge. Under some conditions, the plasma has a high likelihood of quenching, while under other
417 conditions the fuel gas output and coefficient of performance are fluctuating strongly. A thorough
418 understanding of the interrelating physical phenomena in the system will be needed for successful
419 design and optimization of this process. Despite the challenges encountered, the process creates a
420 surplus in energy. More specifically, the chemical energy present in the fuel gas in the outlet gas
421 stream is up to 1.84 times higher than the energy supplied in the form of microwave energy to the
422 gasifier, i.e. a surplus in heating value of up to 84 % is generated with respect to the microwave energy
423 provided. These results give confidence that combining this system with a fuel cell could indeed
424 enable energy recovery, either to form a self-sustained process, or for energy storage and recovery for
425 load leveling purposes in renewable energy production.

426

427 **Acknowledgements**

428 We gratefully thank the Bill and Melinda Gates Foundation (contract OPP1037469) for continuous
429 financial support of the plasma gasification research. We also thank the DEMO team at the P&E
430 department, Verborg Engineering B.V., Euroglass Instruments B.V. (presently Trace Elemental
431 Instruments B.V.), and J.A.S. van Driel for their active technical assistance.

432

433

References

1. Bill & Melinda Gates Foundation, *Water, Sanitation & Hygiene: Reinvent the Toilet Challenge*, Bill & Melinda Gates Foundation 2013.
2. International Energy Agency, *Technology Roadmap Energy Storage*, OECD/IEA 2014.
3. J.A. Turner, A Realizable Renewable Energy Future, *Science* 285, 687–689, 1999.
4. M. Worley and J. Yale, *Biomass Gasification Technology Assessment*, Denver, NREL 2012.
5. T.B. Reed, R. Larson, A wood-gas stove for developing countries, *Energy for Sustainable Development* 3(2), 34–37, 1996.
6. S. Varunkumar, N.K.S. Rajan and H.S. Mukunda, Experimental and computational studies on a gasifier based stove, *Energy Convers. Manage.* 53, 135–141, 2012.
7. E.J.O. Promes, T. Woudstra, L. Schoenmakers, V. Oldenbroek, A. Thallam Thattai and P.V. Aravind, Thermodynamic evaluation and experimental validation of 253MW Integrated Coal Gasification Combined Cycle power plant in Buggenum, Netherlands, *Appl. Energ.* 155, 181–194, 2015.
8. M. Powell, K. Meinhardt, V. Sprenkle , L. Chick and G. McVay, Demonstration of a highly efficient solid oxide fuel cell power system using adiabatic steam reforming and anode gas recirculation, *J. Power Sources* 205, 377–384, 2012.
9. K.D. Panopoulos, L. Fryda, J. Karl, S. Poulou and E. Kakaras, High temperature solid oxide fuel cell integrated with novel allothermal biomass gasification Part II: Exergy analysis, *J. Power Sources* 159, 586–594, 2006.
10. S. Karella, J. Karl and E. Kakaras, An innovative biomass gasification process and its coupling with microturbine and fuel cell systems, *Energy* 33, 284–291, 2008.
11. E. Lorente, M. Millan and N.P. Brandon, Use of gasification syngas in SOFC: Impact of real tar on anode materials, *Int. J. Hydrogen Energ.* 37, 7271–7278, 2012.
12. P.V. Aravind and W. de Jong, Evaluation of high temperature gas cleaning options for biomass gasification product gas for Solid Oxide Fuel Cells, *Prog. Energ. Combust.* 38(6), 737–764, 2012.
13. Ph. Hofmann, K.D. Panopoulos, L.E. Fryda, A. Schweiger, J.P. Ouweltjes and J. Karl, Integrating biomass gasification with solid oxide fuel cells: Effect of real product gas tars, fluctuations and particulates on Ni-GDC anode, *Int. J. Hydrogen Energ.* 33, 2834–2844, 2008.
14. A. Fridman and L.A. Kennedy, *Plasma Physics and Engineering* 2nd ed., Boca Raton, CRC Press 2011.
15. B. Nicolae, Plasma gasification – The waste-to-energy solution for the future, *Problemele Energetice Regionale* 3(26), 107–115, 2014.
16. R. Monteiro Elliott, M.F.M. Nogueira, A.S. Silva Sobrinho, B.A.P. Couto, H.S. Maciel and P.T. Lacava, Tar Reforming under a Microwave Plasma Torch, *Energ. Fuel.* 27, 1174–1181, 2013.
17. K.P. Willis, S. Osada and K.L. Willerton, Plasma Gasification: Lessons learned at Ecovaly WTE facility, *Proceedings of the 18th Annual North American Waste-to-Energy Conference NAWTEC18*, 2010.
18. I.B. Matveev and S.I. Serbin, Modeling of the Coal Gasification Processes in a Hybrid Plasma Torch, *IEEE T. Plasma Sci.* 35(6), 1639–1647, 2007.
19. I.B. Matveev, S.I. Serbin and N.V. Washchilenko, Sewage Sludge-to-Power, *IEEE T. Plasma Sci.* 42(12), 3876–3880, 2014.
20. R. Mourao, R. Ribeiro Marquesi, A.V. Gorbunov, G. Petraconi Filho, A.A. Halinouski and C. Otani, Thermochemical Assessment of Gasification Process Efficiency of Biofuels Industry Waste with Different Plasma Oxidants, *IEEE T. Plasma Sci.* 43(10), 3760–3767, 2015.
21. K. Moustakas, D. Fatta, S. Malamis, K. Haralambous and M. Loizidou, Demonstration plasma gasification/vitrification system for effective hazardous waste treatment, *J. Hazard. Mater.* B123, 120–126, 2005.
22. B. Lemmens, H. Elslander, I. Vanderreydt, K. Peys, L. Diels, M. Oosterlinck and M. Joos, Assessment of plasma gasification of high caloric waste streams, *Waste Manage.* 27, 1562–1569, 2007.
23. Y.C. Hong, D.H. Shin, S.J. Lee, Y.J. Kim, B.J. Lee and H.S. Uhm, Generation of High-Power Torch Plasma by a 915-MHz Microwave System, *IEEE T. Plasma Sci.* 39, 1958–1962, 2011.
24. H.S. Uhm, Y.H. Na, Y.C. Hong, D.H. Shin and C.H. Cho, Production of hydrogen-rich synthetic gas from low-grade coals by microwave steam-plasmas, *Int. J. Hydrogen Energ.* 39(10), 4251–4355, 2014.
25. Y.C. Hong, S.J. Lee, D.H. Shin, Y.J. Kim, B.J. Lee, S.Y. Cho and H.S. Chang, Syngas production from gasification of brown coal in a microwave torch plasma, *Energy* 47, 36–40, 2012.
26. D.H. Shin, Y.C. Hong, S.J. Lee, Y.J. Kim, C.H. Cho, S.H. Ma, S.M. Chun, B.J. Lee and H.S. Uhm, A pure steam microwave plasma torch: Gasification of powdered coal in the plasma, *Surf. Coat. Tech.* 228, S520–S523, 2013.
27. S.J. Yoon and J.G. Lee, Syngas Production from Coal through Microwave Plasma Gasification: Influence of Oxygen, Steam, and Coal Particle Size, *Energ. Fuel.* 26, 524–529, 2012.

- 493 28. S.J. Yoon and J.G. Lee, Hydrogen-rich syngas production through coal and charcoal gasification using
494 microwave steam and air plasma torch, *Int. J. Hydrogen Energ.* 37, 17093–17100, 2012.
495 29. M. Liu, T. Woudstra, E.J.O. Promes, S.Y.G. Restrepo and P.V. Aravind, System development and self-
496 sustainability analysis for upgrading human waste to power, *Energy* 68 377–384, 2014.
497 30. V. Galvita, V.E. Messerle and A.B. Ustimenko, Hydrogen production by coal plasma gasification for
498 fuel cell technology, *Int. J. Hydrogen Energ.* 32, 3899–3906, 2007.
499 31. K. Konno, H. Onodera, K. Murata, K. Onoe and T. Yamaguchi, Comparison of Cellulose
500 Decomposition by Microwave Plasma and Radio Frequency Plasma, *Green Sustainable Chem.* 1, 85–
501 91, 2011.
502 32. Sairem SAS, *Les sources plasma*, 2015, available at <http://www.sairem.fr/les-sources-plasma-56.html>,
503 accessed 30/10/2015, and *Générateurs, composants et mesure*, 2015, available at
504 <http://www.sairem.fr/generateurs-composants-et-mesure-42.html>, accessed 30/10/2015.
505 33. G.S.J. Sturm A.Q. Van Braam Houckgeest, M.D. Verweij, T. Van Gerven, A.I. Stankiewicz and G.D.
506 Stefanidis, Exploration of rectangular waveguides as a basis for microwave enhanced continuous flow
507 chemistries, *Chem. Eng. Sci.* 89 196–205, 2013.
508 34. LabVIEW 2010®, Austin TX: National Instruments Corp., 2010.
509 35. The Engineering Toolbox, *Gross and Net Heating Values for some common Gasses*,
510 http://www.engineeringtoolbox.com/gross-net-heating-values-d_420.html, accessed 09/04/2015.

511