

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/231274123>

In Situ Catalytic Ceramic Candle Filtration for Tar Reforming and Particulate Abatement in a Fluidized-Bed Biomass Gasifier

ARTICLE *in* ENERGY & FUELS · JULY 2009

Impact Factor: 2.79 · DOI: 10.1021/ef900166t

CITATIONS

31

READS

101

6 AUTHORS, INCLUDING:



Sergio Rapagnà

Università degli Studi di Teramo

45 PUBLICATIONS 1,083 CITATIONS

SEE PROFILE



Katia Gallucci

Università degli Studi dell'Aquila

32 PUBLICATIONS 263 CITATIONS

SEE PROFILE



Manuela Di Marcello

Delft University of Technology

20 PUBLICATIONS 196 CITATIONS

SEE PROFILE

In Situ Catalytic Ceramic Candle Filtration for Tar Reforming and Particulate Abatement in a Fluidized-Bed Biomass Gasifier

Sergio Rapagnà,^{*,†} Katia Gallucci,[‡] Manuela Di Marcello,[†] Pier Ugo Foscolo,[‡] Manfred Nacken,[§] and Steffen Heidenreich[§]

Department of Food Science, via C. Lerici 1, 64023 Mosciano, University of Teramo, Italy, Department of Chemistry, Chemical Engineering and Materials, University of L'Aquila, Italy, and Pall Filtersystems GmbH Werk Schumacher, Zur Flügellau 70, 74564 Crailsheim, Germany

Received February 27, 2009. Revised Manuscript Received May 29, 2009

Results are reported for the continuous steam gasification of biomass in a fluidized-bed reactor equipped with a catalytic ceramic candle filter in the freeboard. The filter, a silicon carbide based element (DIA-SCHUMALITH) with a mullite outer membrane, was used as a carrier for a MgO-Al₂O₃-supported Ni catalyst. Tar in the product gas was monitored for gasification runs performed using both catalytic and noncatalytic candle filters over a temperature range of 675–840 °C. It was found that the catalytic candles had the effect of raising the gas yield from 1.35 to 1.55 N m³ per kg of dry biomass and converting 58% of the product tar and 23% of the product methane.

Introduction

Biomass gasification is a thermochemical conversion process that uses combinations of air, oxygen, and steam to produce a fuel gas rich in hydrogen and carbon monoxide and with a significant amount of methane. Carbon dioxide, steam, and nitrogen are also present in the producer gas, as are organic (tar) and inorganic (H₂S, HCl, NH₃, alkali metals) impurities and particulates. The high molecular weight hydrocarbons that constitute the tar are an undesirable and noxious byproduct,¹ the yields of which can be reduced somewhat by careful control of the operating conditions (temperature, biomass heating rate, etc.), appropriate reactor design, and suitable gas conditioning procedures.^{2–4}

Gas cleaning is normally carried out by filtration and scrubbing of the producer gas to reduce its particulate and tar content. In this way, however, the product gas temperature is reduced to close to ambient with a resulting loss of efficiency for subsequent power generation purposes using gas engines: reported values of electric conversion efficiency for such process configurations amount to no more than about 25%, no better than what is obtainable using modern combustion systems coupled with steam turbines. This factor penalizes significantly the overall economic performance for “green” electricity generation based on biomass, for which financial incentives are

offered in many countries. In fact, the complications resulting from the requirement to obtain a tar-free product often contribute significantly to the overall investment and operating costs of small- to medium-scale gasification units. Such systems could provide acceptable local solutions to the energy requirements of many regions, for reasons which include the desirability of establishing distributed power generation as an alternative to socially unacceptable large scale thermal conversion and the reduction in transportation costs that would result from the use of locally produced biomass, a dilute energy source when compared with fossil fuels. Process simplification and intensification have clearly important roles to play in the use of biomass in general and specifically with regard to green gasification-based energy production.

The fact that hot gas cleaning is a focal point of applied research in the gasification field is clear from the number of papers on it appearing in the literature^{5–10} and the number of nationally and internationally funded research projects that deal with it in one way or another. As far as secondary gas treatments

(5) Foscolo P. U., Gallucci K. Integration of particulate abatement, removal of trace elements and tar reforming in one biomass steam gasification reactor yielding high purity syngas for efficient CHP and power plants. Presented at the 16th European Biomass Conference and Exhibition, Valencia, Spain, 2–6 June 2008.

(6) Rapagnà, S.; Mazziotti di Celso, G.; Nacken, M.; Heidenreich, S.; Foscolo, P. U.; Jand, N. Biomass gasification: Hot gas cleaning. Presented at the International Annual Meeting 2007 Catalysis for Environment: Depollution, Renewable Energy and Clean Fuels, Zakopane, September 19–21, 2007.

(7) Heidenreich, S.; Nacken, M.; Salinger, M.; Foscolo, P. U.; Rapagnà, S. Integration of a Catalytic Filter into a Gasifier for Combined Particle Separation and Tar Removal from Biomass Gasification Gas. Presented at GCHT-7 June 23–25, 2008, Newcastle, Australia.

(8) Toledo, J. M.; Corella, J.; Molina, G. Catalytic hot gas cleaning with monoliths in biomass gasification in fluidized beds. 4. Performance of an advanced, second-generation, two-layers-based monolithic reactor. *Ind. Eng. Chem. Res.* **2006**, *45*, 1389–1396.

(9) Diaz-Somoano, M.; Martinez-Tarazona, M. R. Retention of zinc compounds in solid sorbents during hot gas cleaning processes. *Energy Fuels* **2005**, *19*, 442–446.

(10) Delgado, J.; Aznar, M. P.; Corella, J. Biomass gasification with steam in a fluidized bed: Effectiveness of CaO, MgO and CaO-MgO for hot raw gas cleaning. *Ind. Eng. Chem. Res.* **1997**, *36*, 1535–1543.

* To whom correspondence should be addressed. E-mail: rapagna@unite.it.

[†] University of Teramo.

[‡] University of L'Aquila.

[§] Pall Filtersystems GmbH Werk Schumacher.

(1) Bridgwater, A. V. Renewable fuels and chemicals by thermal processing of biomass. *Chem. Eng. J.* **2003**, *91*, 87–102.

(2) Simell, P.; Kurkela, E.; Stahlberg, P.; Hepola, J. Catalytic hot gas cleaning of gasification gas. *Catal. Today* **1996**, *27*, 55–62.

(3) Caballero, M. A.; Corella, J.; Aznar, M. P.; Gil, J. Biomass gasification with air in fluidized bed. Hot gas clean-up with selected commercial and full-size nickel-based catalysts. *Ind. Eng. Chem. Res.* **2000**, *39*, 1143–1154.

(4) van Paasen, S. V. B.; Kiel, J. H. A. Tar formation in a fluidized bed gasifier: Impact of fuel properties and operating conditions; ECN-C-04-013 Report; 2004; pp 1–58.

are concerned, recent advances have included the use of one- or two-layer monolith modules fitted at the gasifier outlet.^{8,11} Although test performances for these modules have been satisfactory, a major drawback for their use on a commercial scale is their rapid deactivation resulting from the decrease in catalyst temperature; to keep this constant throughout the system, reheating of the gaseous stream is often required, at the expense of chemical energy. On the other hand, catalytic filter systems have also been proposed, including a very promising one for coupling to the biomass gasification process.¹²

Work on an effective tar reforming catalytic filter element has been carried out by Pall Filtersystems GmbH Werk Schumacher.^{13,14} This involves the incorporation of a porous inner tube containing a catalyst particle layer in a ceramic hot gas filter candle. An advantage of such catalyst integration is the higher flexibility it provides for potential applications, as the development of an appropriate, highly active catalyst particle system is generally less time-consuming than the alternative of developing a suitable catalytic coating methodology.¹⁴ In addition, a high catalytic capacity can be achieved by simply filling the free hollow cylindrical volume of the filter element with catalyst particles of a high active surface area. Several tar reforming catalysts with different NiO loadings and different catalyst support materials have been tested with synthetic gases, resulting in complete conversion of naphthalene at 800 °C, even in the presence of 100 ppmv of H₂S;¹³ these results suggest that hydrogen sulphide, which is present in producer gas at concentrations of this order, does not cause irreversible deactivation of the catalyst particles in the filter candle, although it certainly reduces its activity.¹⁵

The main incentive for the work described here is the development of an innovative hot gas cleanup technology for the cost-effective production of syngas with the specifications required for use in fuel cells. By integration of a catalytic hot gas filter in the freeboard of a fluidized bed gasifier the thermal efficiency of the whole conversion process is kept high and particle entrainment in the producer gas at the reactor outlet is avoided. The experimental tests of biomass steam gasification reported below were carried out in a reactor system whose main features are shown in Figure 1. Fuel gas quality and tar analyses are reported for each run; the ceramic candle filter (manufactured by Pall Filtersystems GmbH) in the bed freeboard being used both with and without catalyst so that the effect of catalyst incorporation could be assessed.

Experimental Apparatus and Materials

In situ hot gas cleaning was achieved by placing a commercially available ceramic candle filter in the freeboard of a laboratory-scale, fluidized bed gasifier. A sketch of the equipment is shown

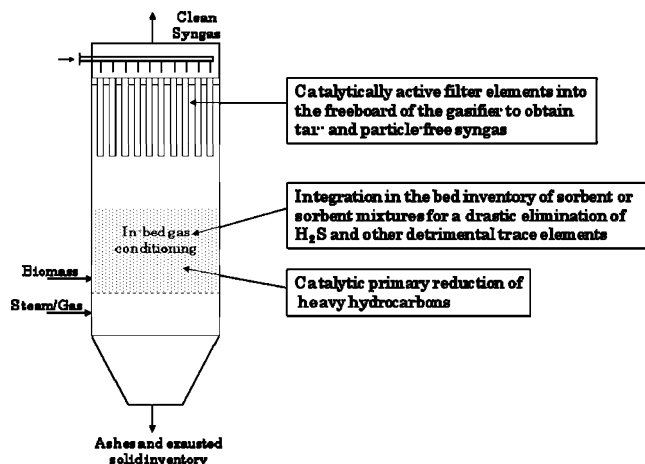


Figure 1. Innovations at the core of UNIQUE project.

in Figure 2. It comprises a bubbling fluidized bed gasifier of internal diameter 0.10 m, heated externally by a 6 kW electric furnace. The biomass is introduced from the top of the bed by means of a continuous feeder unit. A cyclone and a ceramic filter are connected to the gasifier outlet to remove elutriated solid particles, such as ash; although these units become redundant when gas filtration is achieved within the gasifier itself they were left in place to avoid unnecessary system modification. Three condensers in series are used to separate condensable components: the first, of stainless steel, is cooled with tap water and the remaining two, of glass, are cooled with refrigerated diethylene glycol. Online gas analysers (IR and TCD), for the volume composition measurement of H₂, CO, CO₂, CH₄, NH₃, and H₂S, are fed at cold gas rates of 1.5 L min⁻¹ and output to a PC in which the experimental data are stored as functions of time. Temperatures in various plant locations are measured by means of 5 thermocouples, and devices for the continuous monitoring of flow rates and pressures are also employed. Steam was generated from liquid distilled water fed, by means of a peristaltic pump, to a cylindrical, stainless steel evaporator encased in a 2.4 kW electric furnace. The average water flow rate was measured by weighting the feed tank before and after each test; the overall gas flow was measured with a volumetric gasmeter as shown in Figure 2.

The fluidized bed consisted of 3 kg of olivine particles of average diameter 306 μm and density 2500 kg m⁻³. Olivine is known to exhibit a favorable catalytic effect on the gasification process¹⁶ and is more resistant than dolomite to mechanical abrasion. The biomass feedstock consisted of crushed almond shells of chemical composition as reported in Table 1 and an average particle size of 1039 μm. The catalytic ceramic candle filter (a prototype provided by Pall Filtersystems GmbH Werk Schumacher) was fitted in the freeboard of the fluidized bed and equipped on its upstream outer surface with a membrane having an average pore size of 10 μm.

Catalytic Ceramic Filter Preparation. The catalytic activation of the ceramic filter was carried out according to the procedure described for silicon-carbide based filter elements of the type DIA-SCHUMALITH.¹⁴ Briefly, this involved a first impregnation step in which a standard hot gas filter element (of outer diameter 60 mm, inner-diameter 40 mm and length 1520 mm) was impregnated with a fine-wet milled suspension of a MgO-Al₂O₃ precursor using an incipient wetness technique; this was followed by a drying and thermal treatment step to dehydrate the catalyst support coating; in a second impregnation step the support-coated filter element was impregnated with a concentrated nickel nitrate hexahydrate solution to achieve a NiO loading of 60 wt % with respect to the MgO-Al₂O₃ support after drying and final sintering at 900 °C. A 420 mm length of the catalytically activated filter was then cut out from the middle of the treated candle.

(11) Ising M., Gil J., Hunger C. *Gasification of Biomass in a Circulating Fluidized Bed with Special Respect to Tar Reduction*; 1st World Conference on Biomass for Energy and Industry; James & James Science Publishers Ltd.: London, 2001; pp 1775–1778.

(12) Heidenreich S., Nacken M., Foscolo P. U., Rapagnà S., Gasification apparatus and method for generating syngas from gasifiable feedstock materials, PCT/EP2008/003523.

(13) Nacken, M.; Ma, L.; Engelen, K.; Heidenreich, S.; Baron, G. V. Development of a tar reforming catalyst for integration in a ceramic filter element and use in hot gas cleaning. *Ind. Eng. Chem. Res.* **2007**, *46*, 1945–1951.

(14) Nacken, M.; Ma, L.; Heidenreich, S.; Baron, G. V. Performance of a catalytically activated ceramic hot gas filter for catalytic tar removal from biomass gasification gas. *Appl. Catal. B: Environ.* **2009**, *88*, 292–298.

(15) Ma, L.; Verelst, H.; Baron, G. V. Integrated high temperature gas cleaning: Tar removal in biomass gasification with a catalytic filter. *Catal. Today* **2005**, *105*, 729–734.

(16) Rapagnà, S.; Jand, N.; Kiennemann, A.; Foscolo, P. U. Steam-gasification of biomass in a fluidised-bed of olivine particles. *Biomass Bioenergy* **2000**, *19*, 187–197.

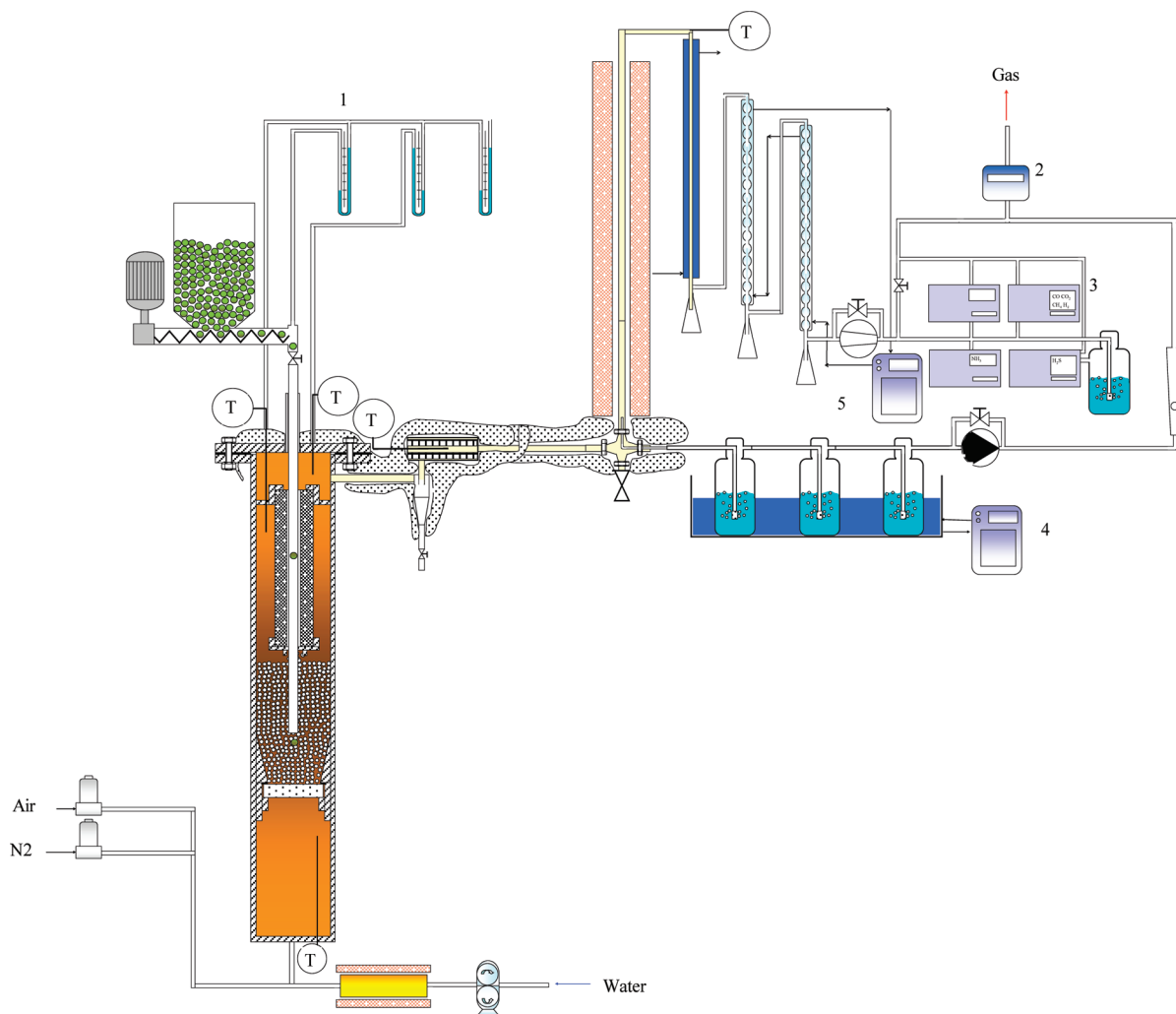


Figure 2. Scheme of the continuous fluidized bed biomass gasification plant: (1) water manometers, (2) gas meter, (3) gas analyzers, (4) thermostat, and (5) chiller.

Table 1. Biomass Chemical Composition

	% wt/wt (as received)	std	% wt/wt (dried at 105 °C)
dry weight	92.30	0.18	
ash			1.19
volatile matter			77.7
fixed carbon			21.1
elemental composition		% wt/wt dry	
C			48.9
H			6.20
N			0.18
O ^a			43.5
Cl			0.029
S			0.026

^a %O = 100 - (%C + %H + %N + %Cl + %S + %ash).

Catalytic Ceramic Filter Characterization. BET Surface. This was measured for 8 mm by 10 mm reference sections of both the catalytically activated filter element and a counterpart noncatalytic element using a QUANTACHROME Autosorb-3 with krypton as the test gas. The measurement error is within 0.005 m²/g under the applied conditions.

Differential Pressure. The differential pressure for the 420 mm lengths of both the catalytically activated and reference filter cylinders was measured at 25 °C as a function of gas face velocity relative to the outer cylinder surface; a Pall Filtersystems GmbH test rig was used for this purpose, the resulting flow resistances being measured in terms of differential pressures with measurement errors within ± 0.2 mbar.

Biomass Gasification Runs. Before a gasification run was started, the gasifier was heated to about 850 °C. For tests with a catalytic candle, a gas mixture of N₂ with 3.5% H₂ was then fed to reduce the NiO present in the catalytic layer to the catalytically active Ni. This was continued until the H₂ content of the outlet gas stream had stabilized at its inlet concentration, at which point the H₂ flow was stopped, the N₂ feed rate decreased to the value reported in Table 2, and steam flow started in preparation for a gasification run.

Biomass feed was started at a rate of 8–10 g/min once the system had stabilized (after 1–2 min of steam flow). This rate was maintained for the entire duration of the run. N₂ was also supplied continuously to assist with the smooth feeding of biomass particles, to stabilize the flow and to adjust the filtration superficial velocity to its predetermined value (of about 90 m/h). Operating conditions for the six gasification runs reported here are shown in Table 2.

The tar content of the producer gas was measured from the condensate using a Shimadzu TOC V-CSN analyzer with the assumption of naphthalene as the model component. When an immiscible organic phase was also obtained following condensation, it was weighted and added to naphthalene measured in the aqueous phase as described above. At the end of each run the amount of char residue in the gasifier was determined by burning it and measuring the total amount of CO and CO₂ released.

Technical specification CEN/TS 15439 provides a standard method for sampling and analyzing the tar and particles in the producer gas. The principle of this method is based on the discontinuous extractive sampling under isokinetic conditions of the gas stream. The sampling train is configured as a heated probe

Table 2. Gasification Operating Conditions of Test

gasification test	I	II	III	IV	V	VI
duration of test (min)	60	60	120	60	60	30
biomass flow rate (g/min)	8	8	8	8	8	10
nitrogen flow rate (L/min)	14	16.5	16.5	16.5	6	0
water supplied (mL)	396	396	808	430	470	400
steam feeding rate (g/min)	6.6	6.6	6.7	7.2	7.8	13.3
filter candle	noncatalytic	noncatalytic	catalytic	catalytic	catalytic	catalytic

Table 3. Characteristics of the Catalytic Filter and Noncatalytic Filter Cylinder

characteristics	catalytic filter cylinder	noncatalytic filter cylinder (reference)
filter type	DIA-SCHUMALITH type N	DIA-SCHUMALITH type N
filter support material (60/40/420 mm)	SiC	SiC
material of filtering outer membrane	mullite	mullite
catalyst support layer	MgO-Al ₂ O ₃ (70–30 wt %)	
catalytically active component	Ni ^a	
BET surface [m ² /g]	0.9	0.013
differential pressure at 25 °C and a face velocity of 90 m/h in air [mbar]	23.8	7.8

^a Deposited as NiO.

with a heated particle filter to remove solid matter. The volatile tars are trapped in heated or chilled impinger bottles (Figure 2) containing 99% isopropanol. Although this facility is available in the gasification test rig, tar measurements presented in this paper are all obtained by means of Shimadzu TOC V-CSN analyzer.

Results and Discussion

Catalytic Ceramic Filter Characteristics. The characteristics of the catalytic and noncatalytic filter cylinders prepared as described above (i.e., filter support, membrane, and catalytic layer material, specific surface area, and differential pressure at 25 °C for various face velocities) are given in Table 3.

The relatively low specific surface of the SiC based filter support of 0.013 m²/g is due to its grain-sintered porous structure. To improve matters, a catalyst support was developed by Pall Filtersystems GmbH Werk Schumacher with which to coat the pore walls of SiC structure to provide a sufficiently large surface over which NiO crystallites could be dispersed.¹⁴ This procedure has resulted in a 69-fold increase in specific surface for the first developed prototype catalytic candle filter (Table 3). However, the integration of the tar reforming catalyst layer on the pore walls leads to a significant increase in flow resistance, resulting in a differential pressure increase from 7.8 to 23.8 mbar at a gas face velocity of 90 m/h. To offset this side effect of integrating the tar reforming catalyst into the filter element the first prepared prototype made use of a much reduced Ni catalyst concentration so as to meet the technical differential pressure requirements for hot gas filtration in this syngas cleanup application.

Ceramic Filter Pressure Drop Tests. A test campaign was carried out to measure the pressure drop as a function of fluid velocity across the filter candle, starting at room temperature and going up to gasification temperature levels. These tests also served to reveal possible gas leaks and to assess the suitability of the system as a whole for high-temperature gasification experiments. For these tests, the reactor was emptied of granular material to ensure a clean filter surface, and air flow rates were measured by means of a mass flow controller (MKS type 1500 Series, flow range 0–50 L/min) and kept constant for about ten minutes. The steady state pressure drop across the filter was measured with two pressure probes inserted in the reactor

through its head, with the tips placed in the regions inside and outside the candle, and connected to a digital manometer (Lab DMM, Bit02, DFP). At room temperature the filtration velocity of 50 m/h could not be exceeded with the mass flow meter. However, the measured data points agree quite well with the trend of the measurements performed by Pall Filtersystems GmbH Werk Schumacher at higher flow rates. When the temperature was increased, higher gas velocities were attainable, exceeding the value of about 100 m/h, which corresponds to that estimated for gasification conditions. These results are reported in Figure 3.

Altogether, the data points at high temperature show a slightly larger pressure drop than the corresponding values at room temperature: the combined increase of gas viscosity and reduction of gas density with temperature may be responsible for this experimental behavior.

The relatively low dependency of pressure drop on operating temperature could also be related to the structure of the filtering medium: the candle is made of two layers in series, and the frictional loss related to the very thin outer mullite layer (with the smallest pore radius) could be assumed as a concentrated source of pressure drop quantifiable in terms of kinetic heads. It could well be the controlling resistance.

It is worth mentioning that, when the reactor has been brought down to room temperature, tests have been repeated with cold air and the results were very much the same as those obtained previously.

Continuous Biomass Gasification Tests. Measurements with and without the catalytically active filter candle integrated in the gasifier were performed. The significant results, such as temperatures, the gas yield, the water conversion, the tar content in the syngas downstream of the catalytic filter element, the char residue, the carbon conversion, the composition of the purified syngas, and the comparison of experimental concentra-

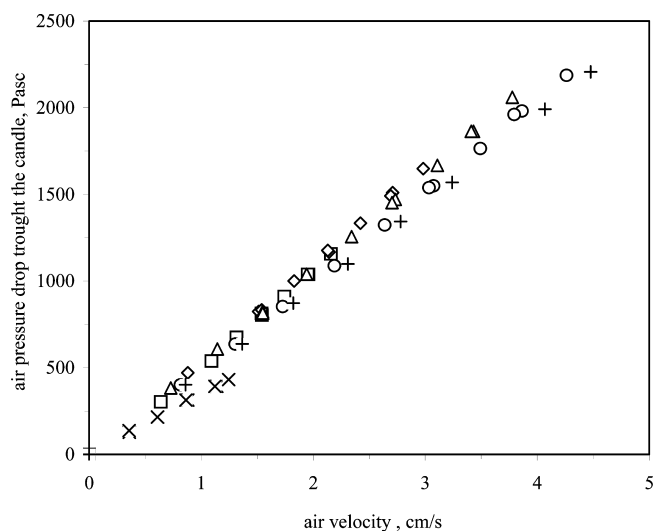


Figure 3. Air pressure drop through the noncatalytic ceramic candle as a function of the external air superficial velocity and reactor temperatures: (x) 19, (□) 263, (◇) 465, (△) 680, (○) 805, and (+) 870 °C.

Table 4. Results of Tests with Filter Candle (I, II) and with a Catalytic Filter Candle (III, IV, V, VI) Installed in the Gasifier

steam gasification of almond shells with olivine bed inventory	I	II	III	IV	V	VI
duration of test (min)	60	60	120	60	60	30
reactor outlet temperature (°C)	675–715	750	791	780	801	801
gasifier bed temperature (°C)	720–803	829	840	832	831	831
steam/biomass dry	0.89	0.89	0.91	0.97	1.06	1.44
water conversion %	15.66	18.64	37.39	26.99	38.92	35.17
gas yield, Nm ³ dry/kg daf (N ₂ free)	0.9–1.1	1.30–1.40	1.60	1.60	1.55	1.80
tar content, g/Nm ³ dry (N ₂ free basis)	6.03	1.90	0.72	not available	0.70	0.92
char residue, g/kg daf	97.39	70.81	81.09	70	54.82 ^a	
carbon conversion %	79.02	85.06	89.00	85.45	88.53	
H ₂ (vol% dry gas, N ₂ free)	34.9–47.1	43.8	48.4	47.8	49.0	50.2
CO ₂ (vol% dry gas, N ₂ free)	21.7	21.0	19.6	20.6	22.1	22.6
CO (vol% dry gas, N ₂ free)	31.6–25.9	25.7	25.8	25.3	22.5	21.3
CH ₄ (vol% dry gas, N ₂ free)	13.4–8.3	9.5	6.2	6.3	6.4	5.9
[H ₂]/[CO ₂]/[CO]/[H ₂ O]	0.30	0.44	0.77	0.64	0.90	0.86
K _p water–gas shift	1.40–1.02	0.94	0.90	0.93	0.93	0.93
filtration velocity (m/h)	85.42	105.72	145.03	109.86	76.63	63.90

^a The total amount of char residue is relative to tests V and VI.

Table 5. Tar Content

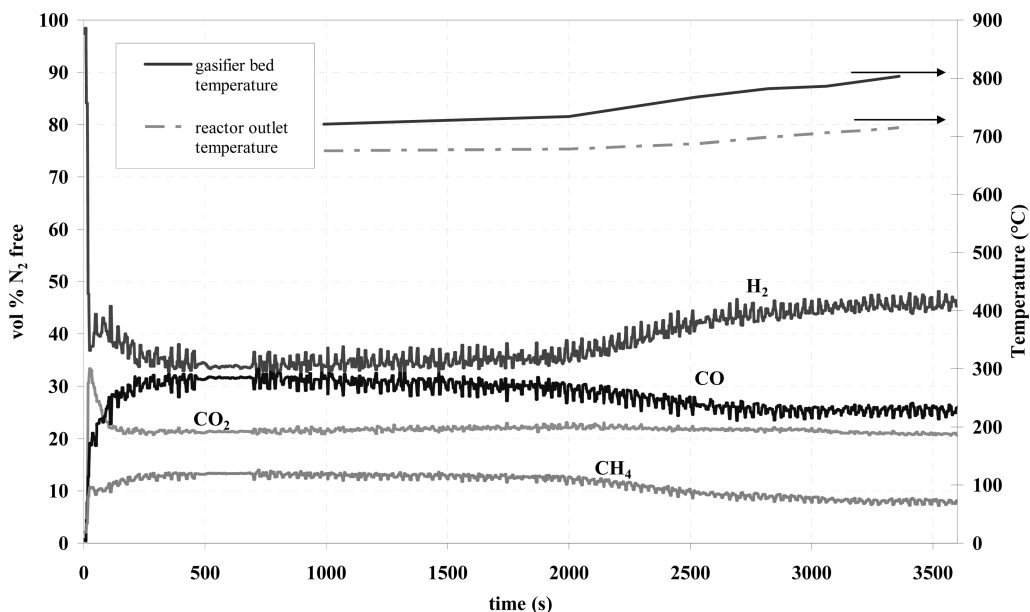
condensate	I	II	III	IV	V	VI
sample 1 (g)	308.65	274.6	415.5	261.00	283.65	253.05
sample 2 (g)	50.10	73	119.85	78.85	26.00	21.25
sample 3 (g)	6.40	4.65	16.8	1.10		
tar content						
TOC sample 1 (mg/L)	5560	2300	1390	2620	1310	1490
TOC sample 2 (mg/L)	9720	3940	2780	not available	2720	3160
TOC sample 3 (mg/L)	11220	not available	3720			

tion ratios with the thermodynamic equilibrium values for the water gas shift reaction, are given in Table 4. Table 5 summarizes the parameters characterizing the condensate collected during each test and the corresponding tar analyses.

Tests V and VI were carried out consecutively, with a jump in the operating conditions as indicated in Table 2. Because of this the combustion of char deposited in the reactor was carried out after test VI so that the quantity reported in Table 4 refers cumulatively to both tests; for mass balance calculation purposes the char residue in each test is assumed proportional to the duration of test. It is clear on comparison of test V with test VI that an increasing ratio of steam to biomass leads to an improvement in gas yield; on the other hand, the water

conversion is slightly reduced, although the number of H₂O moles reacted is greater in absolute terms.

Figure 4 reports significant data for test I, in which the operating temperature was increased progressively with operation time by changing the set point of the electric furnace. A strict relation is found between gasification temperature and composition of the producer gas: the H₂ concentration starts from 35% and reaches 45%; at the same time, the NH₃ concentration in the product gas reduces from 1800 to 1000 vpm. The relatively high tar yield found at the end of this test (6.09 g/Nm³ dry gas yield on average) can be attributed to the low temperature of the bed during the first half of test; this explanation is supported by the result of test II, for which at a

**Figure 4.** Product gas composition and temperature as a function of time (I).

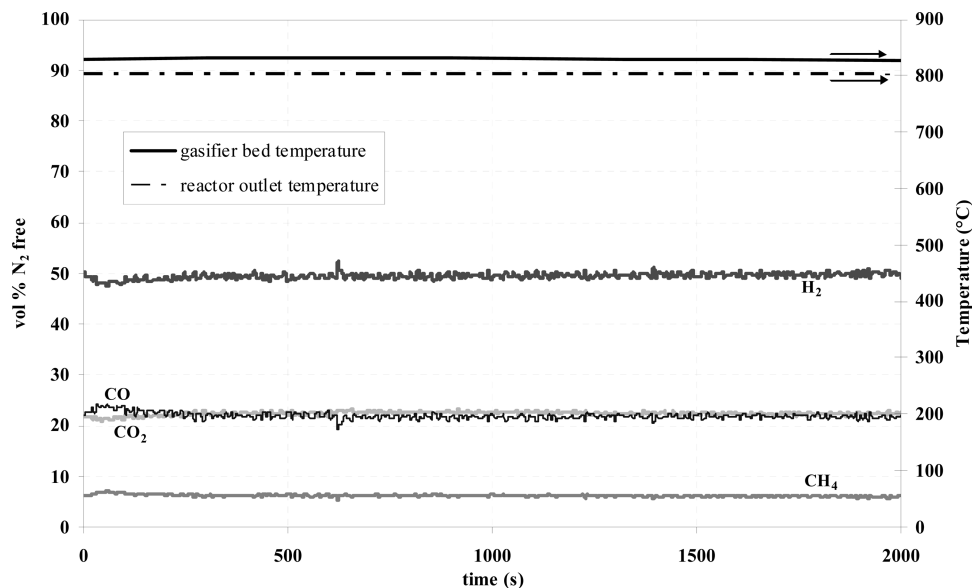


Figure 5. Product gas composition and temperature as a function of time (VI).

constant temperature of 829 °C the measured tar content was only 1.9 g/Nm³.

A significant improvement of all the relevant performance indicators resulted from the use of the catalytic filter candle as reported in Table 4 and 5 with regard to tests III–VI. In these, the reactor temperature remained substantially constant; in test VI, the N₂ flow rate was reduced to zero and at the same time, in order to maintain bubbling fluidization conditions in the bed, the steam feed rate was increased. All this resulted in a high gas yield, high water conversion, low tar content, and close to 50% H₂ concentration by volume in the dry output gas (Figure 5).

The presence of H₂S in the producer gas was also monitored; for the biomass feedstock used in these experiments its concentration remains always well below the concentration used for the activity studies of the catalytic filter candle.¹⁴ The maximum value detected was about 20 ppmv. It is assumed that H₂S deactivation only occurs to a minor extent, because model gas measurements of a filter candle segment in an Al₂O₃ tube reactor with a naphthalene content of 5 g/Nm³ and a 5-fold higher H₂S content at 800 °C show only a decrease of naphthalene conversion from 97% to 58% by H₂S poisoning.¹⁴

The higher gas yield obtained for test VI is mainly caused by an increase in char gasification resulting from the higher water partial pressure in the reactor. As far as the methane and tar content in the product gas are concerned, their amounts are greater than those reported in test III, possibly because of the increased poisoning of the catalytic layer.

Tar conversion for test III (fresh catalyst) shows the same value of 58% as measured under model biomass gasification conditions with 5 g/Nm³ naphthalene in the presence of 100 ppmv H₂S. A possible explanation for this surprising result could be that in this experiment the positive effect of the lower concentration of H₂S in the product gas is countered by the presence of the mixture of hydrocarbons produced during the

biomass devolatilization stage that exhibit reforming rates different from that of the naphthalene model compound.

Conclusions

The innovative concept of integrating a catalytic hot gas filter in the gasifier freeboard has been shown to lead to a simultaneous increase of the gas yield and hydrogen concentration, by significantly reducing the tar content of the raw syngas; at the same time, it also allows efficient particle separation, so that the final result is a hot and clean fuel gas made available right at the exit of the gasifier reactor. When applied to gasification systems of small to medium scale, this would determine a remarkable power plant simplification and intensification, combined with the potential of much higher conversion efficiency of biomass energy to electric power.

Tar conversion extent obtained by means of the catalytic filter is 58%; methane is also partially converted (28%): as a result, a significant increase is obtained in the gas yield (15%) and in its hydrogen concentration, rendering the biomass product gas amenable for fuel cell applications.

Further improvements are expected by the optimization of the catalytic filter system, combining higher catalytic reforming activity with raw gas loading per filter candle at the level of up-to-date commercial standards.

Fluid dynamic studies are also needed, to investigate the fine particle behavior in the reactor freeboard when filter candles are positioned there, the need of a gas pulsing filter cleaning system, the fate of particle clusters detached from the filtration surface, probably floating on the bed surface, and the proper design of a solid withdrawing system from the reactor.

Acknowledgment. The authors would like acknowledge the financial support of the European Commission to the UNIQUE, FP7 European funded project (Grant Agreement number: 211517). M.D.M. is also grateful to the Italian–French University for supporting her PhD grant.

EF900166T