ATMOSPHERIC FLUIDISED BED GASIFICATION OF PROMISING BIOMASS FUELS IN SOUTHERN EUROPEAN REGIONS

by

Kyriakos D. PANOPOULOS, Lydia E. FRYDA, and Emmanuel KAKARAS

Original scientific paper UDC: 662.636/.638-912 BIBLID: 0354-9836, 11, (2007), 1, 5-15

Three promising biomass fuels from southern European regions were gasified atmospherically with air in a lab-scale fluidised bed reactor with quartz or olivine as bed material. The fuels used were an agro-industrial residue (olive bagasse) and the energy crops giant reed and sweet sorghum bagasse. Varying air ratios and temperatures were tested to study the impact on the product gas composition and tar load. Tars were higher in the case of olive bagasse, attributed to its higher lignin content compared to the other two biomasses with higher cellulose. Giant reed gasification causes agglomeration and defluidisation problems at 790 °C while olive bagasse shows the least agglomeration tendency. The particular olivine material promoted the destruction of tars, but to a lesser level than other reported works; this was attributed to its limited iron content. It also promoted the H_2 and CO₂ production while CO content decreased. Methane yield was slightly affected (decreased) with olivine, higher temperatures, and air ratios. Air ratio increase decreased the tar load but at the same time the gas quality deteriorated.

Key words: gasification, fluidised bed, olive, sweet sorghum, giant reed

Introduction

Biomass power based on thermochemical technologies is considered a key option towards reducing greenhouse gas emissions by substituting traditional fossil fuel based generation [1-3]. Sustainable biomass fuels can be: (a) residues from existing agricultural and forestry activities, (b) agro-industrial by-products, (c) the organic fraction of waste materials, such as municipal solid wastes, and (d) energy crops. So far, the technological research and development has been focused on woody fuels from forestry residues and short rotation coppices, which are best options for Northern Europe and the USA. Significant progress has also been gained from the utilization of agro-industrial residues, with most typical fuel example being the olive oil industry residues which is abundant in southern European regions. Numerous publications on olive residues exist; only few are mentioned here [4-8]. The southern European regions also

DOI:10.2298/TSCI0701005P

have a great future potential for implementation of biomass residues as well as annual energy plantations [9], but the reported works on the investigation of thermochemical technologies of these fuels is limited compared to woody fuel options. Since agro-industrial residues are available only in limited periods throughout a year, energy crops could fuel biopower plants during the rest of the time, thus ensuring fuel supply throughout the year. It is therefore important to have comparable experimental data for the behavior of such different biomass fuels under similar conditions in advanced thermochemical processes.

Bubbling and circulating fluidised bed gasifiers have been under significant development over the past decade and appear most suitable for the implementation of biomass in combined heat and power systems in the $+10 \text{ MW}_{th}$ scale applications [1]. Gasification accomplished with air as the gasifying agent at near atmospheric operation is considered the easier technical option [10], which is studied here parametrically for three promising biomass fuels for southern European and Mediterranean regions, namely olive bagasse, sweet sorghum bagasse, and giant reed. The tests were implemented in an experimental lab-scale fluidised bed facility using two different bed materials, namely quartz and olivine which is proven to reduce the tar load in the product gas [11-14].

Olive bagasse is a major agro-industrial residue in Spain, Italy, and Greece. The bagasse from sweet sorghum, which is an annual crop with high photosynthetic efficiency especially in warm conditions with intense sunlight [15], is the by-product from the extraction of the high sugar content of the plant [16] and limited works are reported on its thermochemical utilization [17-18]. Lastly, giant reed (*Arundo donax L.*), is a perennial herbaceous plant, growing wild in south European agro-climatic conditions. Despite being unimproved, wild populations as well as conventional cultivation methods can give high yields of biomass, indicating the great biomass potentiality of this energy crop for the future [19].

Experimental part

Fluidised bed set up and testing procedure

The experimental facility used for the bagasse gasification tests is shown in fig. 1. The reactor is a stainless steel cylindrical tube of 8.9 cm ID and 1.3 m in height, placed in an electrically heated oven for heat loss compensation and preheating. Fluidising air was preheated and introduced into the bed through a perforated type conical distributor. The biomass fuel was delivered with a system of two screw feeders: a first dosimetric followed by one rotating faster and delivering the fuel well into the bed (8 cm above the distributor). A nitrogen overpressure was introduced to the silos to avoid gas back flow. The fluidised bed temperature profile together with several other critical temperatures (freeboard, sampling points, cyclones) were measured using type K chromel-alumel thermocouples with an accuracy of ± 5 °C. The quality of fluidisation was monitored by a differential pressure transducer and all data was displayed and logged on a PC via a data acquisition unit.





(1) – pressurised air flow, (2) – alternative input (e. g. steam), (3) – air/N₂/steam/flue gas mass flow meter, (4) – steam – air valve distributor, (5) – preheater, (6) – distributor, (7) – fluidised bed, (8) – electric resistance, (9) – N₂ flow for backflow prevention, (10, 11) – fuel silo/inverter/electric motor, (12) – freeboard, (13, 14) – primary/secondary cyclone, (15) – ash collection bin, (16) – gas cooler, (17) – condensate collector, (18) – sampling line for gas cleaning, (19) – filter, (20) – product gas destruction system (flare), (21) – heated sampling probe, (22) – heated ceramic filter, (23) – tar measurement, (24) – PC/data collection system

After leaving the freeboard, the product gas was cleaned from coarse particulates in two successive cyclones and was then cooled in a heat exchanger. A cold gas sample stream was further cleaned from particles, tars, and moisture in a gas conditioning unit with a condenser filter and then it was analyzed off-line with gas chromatography. A hot sampling line equipped with a hot ceramic filter (at 350 °C) was delivering gas from the freeboard into a gravimetric tar measurement system, according to the Tar Protocol [20-21], which was composed of a series of six washing bottles containing isopropanol immersed in a salt-ice bath (-20 °C), followed by a membrane pump, a flow and a pressure regulator, and a volume meter to measure total gas sample volume. After approximately 20 minutes of sampling, the tar content was determined gravimetrically, by evaporating the isopropanol solvent under vacuum at 60 °C.

Two sets of gasification experiments were performed using quartz sand and olivine for bed material for each biomass fuel. The chemical composition, density, particle sizes, and amount of the bed materials are given in tab. 1. Olivine occurs abundantly worldwide and consists mainly of a silicate mineral in which magnesium and iron cations are embedded in the silicate tetrahedron. Its catalytic tar cracking capability has been mostly tested as bed inventory in steam gasification experiments [22].

	Quartz	Olivine
Oxide [wt.%]		
- SiO ₂	99	47-49
– FeO	_	7-8
– MgO	_	41-42
- Cr ₂ O ₃ + NiO + CaO + Al ₂ O ₃	~1	<1
Quantity used, [kg]	2	2
Particle sizes, [µm]	125-425	
Material density, [kg/m ³]	2600	3200

Table 1. Bed materials properties

For each bed material type, four air ratios $\lambda = 0.25$, 0.3, 0.35, and 0.40, and two temperature levels (770 and 800 °C) were tested. The steady-states achieved at each of these specified conditions were periods of 120 min. with bed and freeboard temperatures almost constant (standard deviation with time of 3-4% of the average value). Trials with lower temperatures and air ratios were not successful because of increased tar formation which constantly clogged the colder downstream gas path leading to shut down while higher temperature trials of sweet sorghum bagasse and giant reed led to defluidisation of the bed.

Characterization of fuels

The fuels were prepared so as to be fed into the gasifier through a feeding screw. Olive kernel was not further ground; mean particle diameter was about 2-3 mm, as received form the oil extraction units. The sorghum bagasse and cane were crushed and ground into 2-3 mm long stems with a thickness of less than 1 mm. Feeding problems such as cavitations building due to the fuels' stickiness were observed for the two herbaceous fuels (sorghum and cane), which was tackled by adding a vibrating mechanism externally to the fuel silo. The fuels were analyzed for their ultimate, proximate, and ash elemental analysis (tabs. 2 and 3). Furthermore, the composition of the biomass fuels in cellulose, hemicellulose, and lignine was determined using the method of Seaman *et al.* [23]. Acid insoluble lignin content was determined after filtration and drying of acid insoluble solids at 110 C for 24 h and weight correction for ash content. The procedure followed for the non-H₂SO₄-soluble lignin (Klason) is described in ASTM D-1106-84 (Standard Test Method for Acid-Insoluble Lignin in Wood, Reapproved 1990). Acid soluble lignin was determined following TAPPI UM 250 method [24]. The results are shown in tab. 4, where it can be seen that the lignin content of olive bagasse is twice as

Parameter	Olive bagasse	Sweet sorghum bagasse	Giant reed				
Proximate analysis [wt.%]							
Moisture	8.8	8.1	7.4				
Ash	2.4	3.2	2.48				
Ultimate analysis [wt.% dry basis]							
С	51.3	49.5	46.5				
Н	5.8	6.2	5.7				
Ν	1	0.9	0.5				
S	0.0	0.01	0.01				
0*	39	40.1	44.7				
Ash	2.6	3.3	2.6				
HHV d. b. [kJ/kg]**	19 840	18 322	17 980				

Table 2. Proximate and ultimate analysis of the fuels

* by subtraction** higher heating value (HHV) of dry solids (d. b.)

Oxide	Olive bagasse	Sweet sorghum bagasse	Giant reed
SiO ₂	26.4	31.6	44.2
Al ₂ O ₃	4.8	1.9	1.8
Fe ₂ O ₃	7.3	0.4	0.9
CaO	27.1	10.9	1.8
MgO	4.7	6.3	2.8
TiO ₂	0.3	_	0.1
N ₂ O	_	0.2	0.5
K ₂ O	25.8	31.6	30.0
P ₂ O ₅	2.6	3.8	3.2
Cl	_	5.1	_

Table 3. Chemical analysis [wt.%] of the fuels' ash

 Table 4. Main chemical composition of biomass fuels

Diamaga	Composition [wt.% dry ash free basis]		
Biomass	Cellulose	Hemicellulose	Lignin
Olive bagasse	17.6	33.1	49.3
Sweet sorghum bagasse	32.6	44.6	22.8
Giant reed	48.9	47.0	24.9

much as of the other two biomass fuels because of its woody nature, in contrast to the herbaceous nature of the others. In fig. 2, the position of the three fuels is shown on the C-H-O triangle in comparison with coals, pure lignin, and cellulose.



Figure 2. Composition of fuels used in comparison with coals, pure lignin, and cellulose in a C-H-O triangular diagram

Results and discussion

The dry product gas composition graphs from olive bagasse, sweet sorghum bagasse and giant reed gasification tests are shown in figs. 3, 4, and 5, respectively. For olive and sweet sorghum bagasse two graphs are presented: the left hand side at 770 °C and the right hand side at 800 °C (average temperatures). In the case of giant reed gasification, the bed defluidised quickly during the 800 °C test for both bed materials and further tests at this temperature could not be included in the test matrix (fig. 6). In the graphs of figs. 3, 4, and 5, the measured tar content is also plotted in the form of column bars. In all cases, the dashed lines correspond to the quartz bed tests while solid lines are used for olivine tests.

From the results it can be concluded that olivine promotes H_2 and CO_2 formation and lowers CO production. For similar air ratios, the methane yield appears slightly lower in the olivine bed. Furthermore, olivine drops more significantly the amount of tars in the

Panopoulos, K. D., Fryda, L. E., Kakaras, E.: Atmospheric Fluidised Bed Gasification of ...



Figure 3. Gas composition (lines) and tar (bars) content of olive bagasse gasification tests at (a) 770 °C and (b) 800 °C in quartz (dashed) and olivine bed (solid) vs. air ratio



Figure 4. Gas composition (lines) and tar (bars) content of sweet sorghum bagasse gasification tests at (a) 770 °C and (b) 800 °C in quartz (dashed) and olivine bed (solid) vs. air ratio

gas in all cases, but at lower rates than reported in other research works published [11, 12, 20]. This can be attributed to the fact that the particular olivine used did not have a very high iron content (tab. 1), which is considered a key to the catalytic tar cracking ability of olivine [25].

Gasifier operation at 800 °C produces slightly less tar amounts than at 770 °C. Tar amounts drop sharply with higher air ratios, but so does the quality of the product gas (H₂ and CO drop in favor of CO₂ and H₂O). Olive bagasse gasification produces higher tar contents than the other two fuels for gasification parameters 770 °C and 0.25 air ratio; this can be attributed to the higher lignin content of this particular fuel (tab. 4). Indeed other experimental works show that lignin and xylan are more difficulty gasified while cellulose is completely converted at the early steps of the process [26]. Above that temperature and air ratio though, no conclusion can be drawn for the tar content as the oxidation environment seems to have a much stronger effect on tar destruction.



THERMAL SCIENCE: Vol. 11 (2007), No. 1, pp. 5-15

Feed interruption or silo recharge

60

40 요

30

Defluidisation

1000

900

800

700



Figure 5. Gas composition (lines) and tar (bars) content of giant reed gasification tests at 770 °C in quartz (dashed) and olivine bed (solid) vs. air ratio

Figure 6. Bed pressure drop and temperature profile during the gasification test of giant reed in an olivine bed which led to defluidisation

During the experiments the loss of fluidisation was indicated by the sudden pressure drop and by the loss of temperature uniformity across the bed (fig. 6). During the giant reed tests the bed particles agglomerated due to the fuels' higher potassium content (tab. 3) which led to the formation of eutectic ash components probably consisting of potassium silicates; these produced a viscous melt at temperatures below 800 °C and covered the bed particles with sticky melt acting as glue between them [27-29]. There are several reported works on ash related sintering and agglomeration in fluidized bed reactors (FBs) performing combustion or gasification. According to Skrifvars et al. [30], three sintering mechanisms are identified in solid fuel FBs: (a) partial ash melting, (b) partial melting with the formation of a viscous liquid (viscous flow sintering), and (c) chemical reaction forming a layer of a new compound on particles (desulphurisation processes, when CaO from the ash or from added dolomite and gaseous SO₂ are present). The dominant mechanism depends mainly on the chemical and mineralogical composition of the ash involved. Viscous flow sintering mechanism occurs in silicate systems especially with alkali rich fuels such as biomass. Ash derived alkalis deposit on the bed material or silicate ash particles as (1) small particles, (2) condensation of alkali species (KCl, KOH, K₂SO₄, K), or (3) chemical reaction with particle surface followed by homogenization and strengthening of the inner layer of the coating that finally leads to melting and adhesion increase, responsible for the agglomeration result [29, 31, 32].

By performing some extra sweet sorghum tests at slightly higher temperatures than 800 °C it was observed that this fuel also led to defluidisation (at approx. 810 °C). On the contrary, olive bagasse did not create similar problems even at temperatures above 830 °C. This was attributed to its much higher Fe content in comparison to the other two fuels, and to a lesser extent to their Ca and Mg ash content, which all shift the ash melting temperatures higher. A detailed analysis on the agglomeration behavior of the tested fuels will be published in the near future.

Conclusions

This work aimed at the experimental investigation of atmospheric fluidised bed gasification of promising residues and annual energy crops from southern European regions, using air as gasification medium, which is the simplest and most cost effective technical option. Giant reed caused agglomeration problems in both quartz and olivine beds leading to defluidisation at temperatures less than 800 °C. Olive bagasse turned out as the most agglomeration resistant fuel of the three tested. The gasification of olive bagasse produced larger tar quantities at the lower temperatures (770 °C) and the smaller air ratio (0.25) compared to the other two fuels, probably because of its higher lignin content.

The particular olivine used did not have a very high iron content and its tar destruction capability was therefore limited. Nevertheless, the measured tar loads in all cases were lower compared to the corresponding tests in the quartz bed. Olivine affected the permanent product gas composition as well, by promoting H_2/CO_2 and slightly lowering CO content. Methane yield was reduced slightly but remained almost constant for all experiments. Increasing air ratios significantly reduced the amount of tars but at the same time the gas quality as well by increasing CO_2 and H_2O (in the expense of fuel content such as CO and H_2).

References

- Maniatis, K., Millich, E., Energy from Biomass and Waste: The Contribution of Utility Scale Biomass Gasification Plants, *Biomass and Bioenergy*, 15 (1998), 3, pp. 195-200
- Spliethoff, H., Status of Biomass Gasification for Power Production, *IFRF Combustion Journal 2001*, Article number 200109
- [3] Bridgwater, A. V., The Future for Biomass Pyrolysis and Gasification: Status, Opportunities and Policies for Europe, ALTENER Contract No: 4.1030/S/01-009/2001, Bio-Energy Research Group, 2002, Ashton University, Birmingham, UK
- [4] Andre, R. N., Pinto, F., Franco, C., Dias, M., Gulyrtlu, I., Matos, M. A. A., et al., Fluidised Bed Co-Gasification of Coal and Olive Oil Industry Wastes, Fuel, 84 (2005), 12-13, pp. 1635-1644
- [5] Garcia-Ibanez, P., Cabanillas, A., Sanchez, J. M., Gasification of Leached Orujillo (Olive Oil Waste) in a Pilot Plant Circulating Fluidised Bed Reactor, Preliminary Results, *Biomass and Bioenergy*, 27 (2004), 2, pp. 183-194
- [6] Roig, A., Cayuela, M. L., Sánchez-Monedero, M. A., An Overview on Olive Mill Wastes and Their Valorisation Methods, *Waste Management*, 26 (2006), 9, pp. 960-969
- [7] Ollero, P., Serrera, A., Arjona, R., Alcantarilla S., The CO₂ Gasification Kinetics of Olive Residue, *Biomass and Bioenergy*, 24 (2003), 2, pp. 151-161
- [8] Encinar, J. M., Beltrán, F. J., Ramiro, A., González, J. F., Pyrolysis/Gasification of Agricultural Residues by Carbon Dioxide in the Presence of Different Additives: Influence of Variables, *Fuel Processing Technology*, 55 (1998), 3, pp. 219-233
- [9] Jankes, G. G., Milovanović, N. M., Biomass Gasification in Small-Scale Units for the Use in Agriculture and Forestry in Serbia, *Thermal Science*, 5 (2001), 2, pp. 49-57
- [10] Bridgwater, A. V., The Technical and Economic Feasibility of Biomass Gasification for Power Generation, *Fuel*, 74 (1995), 5, pp. 631-653

- [11] Devi, L., Craje, M., Thüne, P., Ptasinski, K. J., Janssen, F. J. J. G., Olivine as Tar Removal Catalyst for Biomass Gasifiers: Catalyst Characterization, *Applied Catalysis A 294* (2005), 1, pp. 68-79
- [12] Devi, L., Ptasinski, K. J., Janssen, F. J. J. G., van Paasen, S. V. B., Bergman, P. C. A., Kiel, J. H. A., Catalytic Decomposition of Biomass Tars: Use of Dolomite and Untreated Olivine, *Renewable Energy*, 30 (2005), 4, pp. 565-587
- [13] Guan, H., Shaoping, X., Shiguang, L., Changrui, X., Shuqin, L., Steam Gasification of Apricot Stones with Olivine and Dolomite as Downstream Catalysts, *Fuel Processing Technol*ogy, 87 (2006), 5, pp. 375-382
- [14] Rapagnà, S., Jand, N., Kiennemann, A., Foscolo, P. U., Steam-Gasification of Biomass in a Fluidised-Bed of Olivine Particles, *Biomass and Bioenergy*, 19 (2000), 3, pp. 187-197
- [15] Venturi, P., Venturi, G., Analysis of Energy Comparison for Crops in European Agricultural Systems, *Biomass and Bioenergy*, 25 (2003), 3, pp. 235-255
- [16] Suresh, K., Kiran Sree, N., Venkateswer Rao, L., Utilization of Damaged Sorghum and Rice Grains for Ethanol Production by Simultaneous Saccharification and Fermentation, *Bioresource Technology*, 68 (1999), 3, pp. 301-304
- [17] Handan Çubuk, M., Heperkan Hasan, A., Investigation of Pollutant Formation of Sweet Sorghum-Lignite (Orhaneli) Mixtures in Fluidised Beds, *Biomass and Bioenergy*, 27 (2004), 3, pp. 277-287
- [18] Piskorz, J., Majerski, P., Radlein, D., Scott, D. S., Bridgwater, A. V., Fast Pyrolysis of Sweet Sorghum and Sweet Sorghum Bagasse, *Journal of Analytical and Applied Pyrolysis, 46* (1998), 1, pp. 15-29
- [19] Cosentino, S. L., Copani, V., D'Agosta, G. M., Sanzone, E., Mantineo, M., First Results on Evaluation of Arundo Donax L. Clones Collected in Southern Italy, Industrial Crops and Products, 23 (2006), 2, pp. 212-222
- [20] Neeft, J. P. A., Knoef, H. A. M., Zielke, U., Sjöström, K., Hasler, P., Simell, P. A., et. al., Guideline for Sampling and Analysis of Tar and Particles in Biomass Producer Gases, Version 3.3, Energy project ERK6-CT1999-20002 (Tar Protocol)
- [21] Maniatis, K., Beenackers, A. A. C. M., Tar Protocols. IEA Bioenergy Gasification Task, *Biomass and Bioenergy*, 18 (2000), 1, pp.1-4
- [22] Rapagna, S., Jana, N., Kiennemann, A., Foscolo, P. U., Steam-Gasification of Biomass in a Fluidised-Bed of Olivine Particles, *Biomass and Bioenergy*, 19 (2000), 3, pp. 187-197
- [23] Seaman, J. F., Bubl, J. L., Harris, E. E., Quantitative Saccharification of Wood and Cellulose, Ind. Eng. Chem. Anal. Ed., 17 (1945), pp. 35-37
- [24] ***, TAPPI Test Method T250, Acid-Soluble Lignin in Wood and Pulp., in: Tappi Test Methods, Technical Association of the Pulp and Paper Industry, Atlanta, Ga., USA
- [25] Nordgreen, T., Liliedahl, T., Sjöström, K., Metallic Iron as a Tar Breakdown Catalyst Related to Atmospheric, Fluidised Bed Gasification of Biomass, *Fuel*, 85 (2006), 5-6, pp. 689-694
- [26] Hanaoka, T., Inoue, S., Uno, S., Ogi, T., Minowa, T., Effect of Woody Biomass Components on Air-Steam Gasification, *Biomass and Bioenergy*, 28 (2005), 1, pp. 69-76
- [27] Öhman, M., Experimental Studies on Bed Agglomeration during Fluidised Bed Combustion of Biomass Fuels, Ph. D. thesis, Energy Technology Centre, Piteå, Sweden, Department of Chemistry, Inorganic Chemistry, Umeå University, Sweden, 1999
- [28] Skrifvars, B.-J., Öhman, M., Nordin, A., Hupa, M., Predicting Bed Agglomeration Tendencies for Biomass Fuels Fired in FBC Boilers: A Comparison of Three Different Prediction Methods, *Energy & Fuels*, 13 (1999), 2, pp. 359-363
- [29] Van der Drift, A., Olsen, A., Conversion of Biomass, Prediction and Solution Methods for Ash Agglomeration and Related Problems, Final report, ECN Biomass, ECN-C-99-090, 1999
- [30] Skrifvars, B. -J., Hupa, M., Backman, R., Hiltunen, M., Sintering Mechanisms of FBC Ashes, Fuel, 73 (1994), 2, pp. 171-176
- [31] Ohman, M., Nordin, A., Bed Agglomeration Characteristics During Fluidised Bed Combustion of Biomass Fuels, *Energy & Fuels*, 14 (2000), 1, pp. 169-178

Panopoulos, K. D., Fryda, L. E., Kakaras, E.: Atmospheric Fluidised Bed Gasification of ...

[32] Baxter, L., Miles, T. R., Miles Jr, T. R., Jenkins, B. M., Milne, T., Dayton, D., Bryers, R. W., Oden, L. L., The Behavior of Inorganic Material in Biomass-Fired Power Boilers: Field and Laboratory Experiences, *Fuel Processing Technology*, 54 (1998), 1-3, pp. 47-78

Authors' address:

K. D. Panopoulos, L. E. Fryda, E. Kakaras

National Technical University of Athens, School of Mechanical Engineering, Thermal Engineering Section, Laboratory of Steam Boilers and Thermal Plants Building "O" 9, Heroon Polytechniou ave., Zografou GR 15780, Athens, Greece

Corresponding author L. E. Fryda E-mail: lfryda@central.ntua.gr

Paper submitted: August 31, 2006 Paper revised: February 21, 2006 Paper accepted: February 25, 2006