ECN

Energy research Centre of the Netherlands

Experimental results from the allothermal biomass gasifier Milena

C.M. van der Meijden

A. van der Drift

B.J. Vreugdenhil

Presented at the 15th European Biomass Conference, 7-11 May 2007, Berlin, Germany

ECN-M--08-083 November 2008

EXPERIMENTAL RESULTS FROM THE ALLOTHERMAL BIOMASS GASIFIER MILENA

C.M. van der Meijden, A. van der Drift, B.J. Vreugdenhil
ECN Biomass, Coal and Environmental Research, PO Box 1, 1755 ZG Petten, the Netherlands

ABSTRACT: ECN is developing an indirectly heated (allothermal) biomass gasification process (Milena), optimized for the production of Synthetic Natural Gas (SNG). The methane rich gas from the gasifier is cleaned and the carbon monoxide and hydrogen in the gas are converted into methane by a catalyst. After removal of carbon dioxide and water pure methane is available to inject into the natural gas grid. Some operational parameters, like gasifier temperature and bed material, have influence on the gas composition and therefore on the overall efficiency to SNG. Experiments in a 30 kW_{th} Milena gasifier were done to determine the optimal operating conditions for SNG production. The results of these tests are presented in this paper.
Keywords: allothermal conversion, biomass conversion, fluidized bed, gasification, methane, tar.

1 INTRODUCTION

Natural gas is an important energy source in the Netherlands. Natural gas is the most convenient and environmental friendly energy source for central heating.

As the natural gas reserves are finite and contribute to CO_2 emissions a sustainable alternative is required to replace current fossil based gas supplies. This substitute gas is to be considered as a secondary energy carrier as the primary source will necessarily be based on renewable biomass derived resources.

Gasification in combination with methanation of the produced gas is a possible route to produce this sustainable Substitute Natural Gas (SNG).

ECN (Energy research Centre of the Netherlands) is developing an indirectly heated (allothermal) biomass gasification process (Milena), optimized for the production of SNG. In the Milena gasifier, biomass is gasified in a riser, which is heated by circulating sand from a fluidized bed combustor. The combustor is fed with the remaining biomass char from the gasification section. The gasifier / riser and the fluidized bed combustor are integrated in a single vessel to make pressurization of the process easy.

For the efficient production of SNG, a producer gas with a high concentration of hydrocarbons is ideal. This requires a gasification process at a low temperature (below 1000°C). A low temperature air blown allothermal gasification process can produce SNG from biomass with an overall efficiency of more than 70% [1].

Low temperature gasifiers produce a considerable amount of tar. The tar content in the producer gas can be reduced by a catalyst in or after the gasifier. In general these catalysts require addition of extra steam to the gasifier to prevent the formation of soot. The additional steam reduces the overall efficiency of the process.

ECN has chosen not to reduce the tar content catalytic, but to remove the tar from the gas and to use the tar as fuel for the combustion section of the gasifier. The selected tar removal technology is the Olga technology [2], which was jointly developed by ECN and Technisch Bureau Dahlman and currently commercially available.

For the further development and scale up of the technology experimental data is required. Here, we present results from experiments carried out in the lab-scale ECN Milena gasifier.

2 EXPERIMENTAL LAY-OUT

2.1 Milena lab-scale gasifier

ECN realized a 30 kW_{th} lab-scale Milena gasifier in 2004, capable of producing approximately 8 m_n^3/h methane-rich medium calorific gas with high efficiency. Figure 1 shows schematically the working principles of the lab-scale installation.

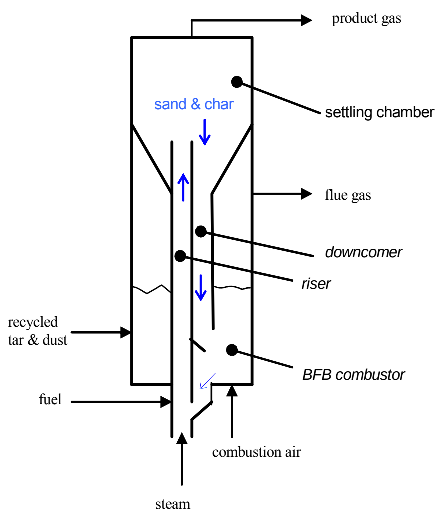

Figure 1: Schematic representation of Milena lab-scale gasifier, arrows represent the solids flow

Biomass (typical 6 kg/h) is fed into the riser (36 mm i.d.) were a small amount of superheated steam is added (typical between 0.1 and 2 kg/h). Hot bed material (typical sand or olivine 0.2 – 0.3 mm at 925°C) enters the riser from the combustor through a hole in the riser, which is located under the biomass feeding point.. The bed material heats the biomass to typical 850°C. The heated fuel particles degasify and create a vertical linear velocity of approximately 6 m/s, creating a “turbulent fluidization” regime in the riser and a carry over of the bed material together with the degasified biomass particles (char). The vertical velocity of the gas is reduced in the settling chamber, causing the larger solids (bed material and char) to separate from the gas and fall down into the downcomer. The producer gas leaves the reactor from the top and is sent to the cooling and gas cleaning section. The typical residence time of the gas is between 1 and 2 seconds.

The char is combusted in the bubbling fluidized bed, were it heats the bed material to approximately 925°C. The CO and C_xH_y in the flue gas is reduced to a relatively low level in the freeboard by adding secondary air. Flue gas leaves the reactor and is cooled and de-dusted before it is sent to the stack. The heated bed material leaves the bubbling fluidized bed from the bottom and is sent to the riser again.

The lab-scale installation is made of stainless steel (grade 253MA). Heat loss from the process is compensated by high temperature electrical trace heating and external insulation.

The temperature in the gasifier is measured at different locations. A thermocouple in the gas inlet nozzle is used for operational purposes only, because the fluidization gas cools the thermocouple. The gasifier outlet temperature is measured in the settling chamber and in the gas outlet. The temperature measured in the settling chamber is seen as the most accurate gas outlet temperature measurement, because the thermocouple is in direct contact with sand in the settling chamber. The combustor temperature is measured by 4 different thermocouples in the bubbling fluidized bed.

The concentration of CO, H_2, CO_2, CH_4, C_2H_2, C_2H_4, C_2H_6, C_6H_6, C_7H_8, Ar and N_2 are measured on-line every minute with gas monitors and/or a micro gas chromatograph. The concentration of tar in the gas is measured using the SPA method [3].

Part of the producer gas (2 m_n^3/h) is cleaned in the Olga tar removal unit and sent to the methanation unit. The remaining gas is combusted. Results from the methanation experiments will be presented in the future.

2.1 Test conditions

The tests described here were carried out between May 2004 and March 2007. The standard fuel for the tests was dry beech wood delivered by Rettenmaier from the Netherlands, the composition given in table I is an average of two analyses.

Table I: Composition of wood used in Milena gasifier

	Clean wood
Moisture (wt% ar)	9.0
Ash (wt% db)	1.0
C (wt% daf)	49.2
H (wt% daf)	6.2
O (wt% daf)	44.4
N (wt% daf)	0.2
S (wt% daf)	0.03
Cl (wt% daf)	0.004
HHV (MJ/kg daf)	19.8
Sieve fraction (mm)	0.75 – 3

The wood particles used in the lab-scale installation are relatively small, because the dimensions of the feeding system, the riser and the downcomer are small. Larger particles will cause operational problem.

The Milena gasifier was always heated to process temperature prior to the test by the electrical trace heating and combustion of methane in the combustor.

Most of the bed material is in the combustor. During the nights between tests the bed material was purged with air and kept at 800 – 900°C.

The combustor was operated at a typical air to fuel ratio of 1.25. Methane was added to the combustor to simulate a tar recycle from the Olga gas cleaning section. Adding real tar was successfully tested, but addition of methane was more practical.

The bed materials used in the reactor were sand and olivine (see table II). Olivine was selected because of the positive results in respect of tar reduction produced with the FICFB gasifier in Güssing [4].

The olivine was greenish before the test and turned red when it was used. The olivine was used for more than 100 gasification hours and was kept above 850°C for several weeks to activate the olivine.

Table II: Bed materials used in Milena gasifier

	Quartz sand	Olivine
Origin	-	Norway
Sieve fraction [mm]	0.1 - 0.5	0.06 – 0.25
Density [kg/m^3]	2600	3250

The variation in temperature was limited during the tests with olivine as a bed material, because the original goal of the tests was to reduce the tar content in the gas, therefore a relative high gasification temperature was selected.

None of the tests with beech wood resulted in an unwanted stop, caused by agglomeration of the bed, so the bed materials could be used again. The loss of bed material was compensated by adding fresh bed material to the combustor. The bed level in the combustor was kept at approximately 50 cm above the bed plate.

The duration test described here was the first test were the Milena producer gas was cleaned by the Olga gas cleaning and sent to the methanation test unit after Cl en S removal by adsorbentia. Because of the limited experience with the integrated SNG system the process conditions were chosen to minimize operational problems. The concentration of tar in the producer was reduced by using olivine as bed material. The steam to biomass ratio was 0.28 to minimize possible soot formation problems in the methanation unit.

3 RESULTS

The data produced during different tests and for different projects were analyzed. Component balances were made to check consistency of the measurement data. Reliable tests were selected and are reported here.

3.1 Effect of process conditions on gas composition

Most catalytic active bed materials used for tar reforming also promote the CO shift reaction.

$$CO + H_2O \leftrightarrow H_2 + CO_2 \quad (1)$$

The theoretical CO shift equilibrium temperatures for the different test were calculated from the measured concentrations of CO, H_2 and CO_2 and the calculated H_2O concentration for each test. The differences between the calculated CO shift equilibrium temperatures and the process temperatures (ΔT_{shift}) are given in figure 2.

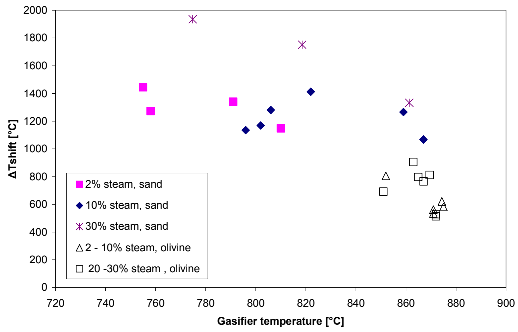

Figure 2: Effect of temperature on CO shift equilibrium

Normally the CO shift equilibrium concentration is not reached in the Milena lab-scale gasifier. The H_2 concentration is below the theoretical equilibrium concentration. This is also observed in other ECN fluidized bed gasifiers where not a catalytic active bed material is used and the accumulation of char is limited.

The offset from equilibrium temperature decreases by higher temperatures. The use of olivine instead of sand also promotes the CO shift reactions (more H_2 production). The addition of steam increases the amount of hydrogen in the producer gas, but has no clear effect on the calculated equilibrium temperature.

A high methane yield improves efficiency to SNG or electricity via a gas turbine or gas engine. Figure 3 shows the relation between gasifier temperature and methane yield for different bed materials and steam to biomass ratios.

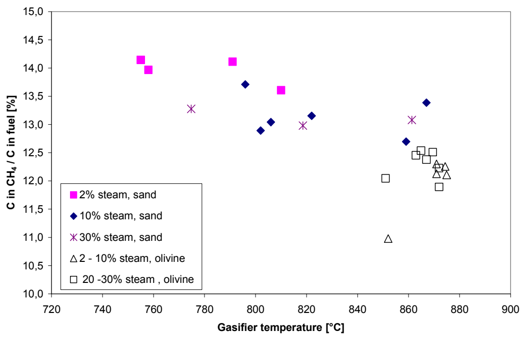

Figure 3: Effect of temperature on CH_4 production

There is a weak dependency of methane yield on gasifier temperature. A low gasification temperature is in favor of the methane yield, but 750°C is seen as a lower limit for the operation of the Milena gasifier, due to lack of experience with the behavior of tar produced at low gasification temperatures in the Olga gas cleaning. The replacement of sand by olivine decreases the amount of methane produced.

The olivine test at 852°C and a low amount of fluidization steam resulted in a relatively low methane yield (lowest point in figure 3), the reason is not clear.

A potential interesting product from biomass is ethylene. Ethylene is a base chemical for the production of plastics. The economic value of ethylene is higher than the value of methane. The measured ethylene yields for the different tests are shown in figure 4.

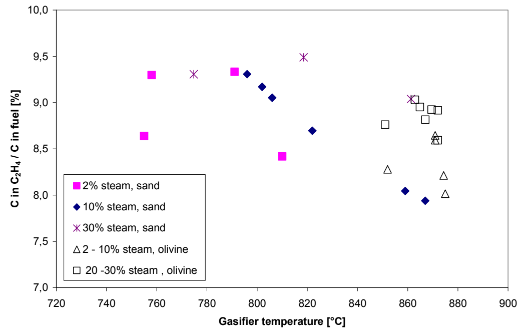

Figure 4: Effect of temperature on C_2H_4 production

The tests using sand as a bed material and a steam to biomass ratio of 10% show a clear relation between temperature and ethylene yield. This relation is not confirmed by the other tests using sand as a bed material. The olivine tests show a weak dependency on temperature. The olivine used for the test described here does not reduce the ethylene yield.

The amount of tar in the producer gas is high compared to the FICFB process [4], staged gasifiers or downdraft gasifiers. The Milena process design is not optimized towards tar reduction, because the Olga tar removal process is used to remove the tar. Figure 5 shows the measured tar yields for the different tests.

The yield of tar decreases with increasing temperature. The effect of olivine on tar reduction is limited.

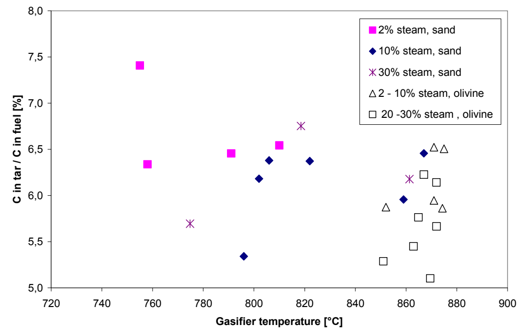

Figure 5: Effect of temperature on tar production

Others have measured much lower tar yields when olivine was used as a bed material in a bubbling fluidized bed reactor at relatively high steam to biomass ratios of 0.6 – 0.7 [5]. The residence time of the gas in the Milena riser and the settling chamber is relatively short (1 – 2 seconds), the solids concentration is lower than in a bubbling fluidized bed and the water concentration in the producer gas is kept relatively low to keep the overall efficiency high. These conditions are not in favor of tar reduction.

Figure 6 shows the calculated carbon conversion in the riser. The carbon conversion is defined as amount of carbon in the producer gas (including tar, excluding dust in the gas) divided by the amount of carbon in the fuel. There is a weak dependency of carbon conversion on gasifier temperature. The carbon conversion is heavily influenced by the particle size of the fuel. The relatively small particles give a high carbon conversion. It is expected that carbon conversion will drop when larger fuel particles will be used during the experiements in the pilot plant which is under construction at the moment.

Carbon not converted into gas is char. More than 98% of the char is removed from the producer gas in the settling chamber and is recycled to the combustor. The relatively high collection efficiency of the settling chamber is caused by the absence of dust in the fuel. Other fuels (grass) resulted in lower collection efficiencies. The char is completely combusted in the fluidized bed combustor. The ash leaving the combustor is free of carbon.

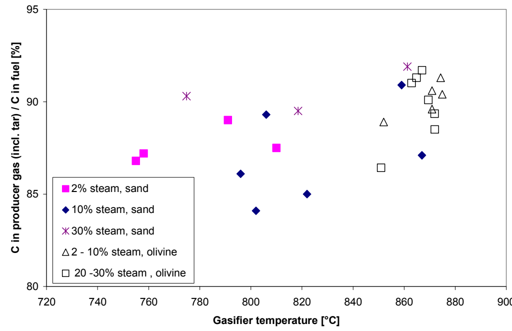

Figure 6: Effect of temperature on carbon conversion

The char not sent to the combustor ends up in the producer gas and is removed from the producer gas in the gas cleaning section; this char stream will also be sent to the combustor in commercial applications.

3.2 Duration test

The primary goal of the duration test was to deliver clean gas to the methanation unit; this goal was achieved without any interruption. Figure 7 shows the variation in producer gas composition direct after the Milena gasifier. The data shown is raw measurement data. The measurements were interrupted because of the required cleaning of the gas sampling system. The periodic disruptions were caused by switching between the two fuel bunkers for refilling.

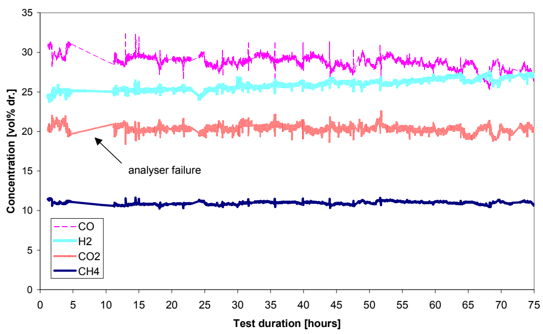

Figure 7: Raw gas composition Milena outlet

The hydrogen and carbon monoxide concentrations were not stable during the test period. The hydrogen concentration was increasing and the carbon monoxide concentration was decreasing during the test. A possible explanation is an increased catalytic activity of olivine promoting the CO shift and reform reactions. The tar concentration in the gas decreased from 31 to 24 g/m_n^3, over the test period.

Table III gives the measured average gas composition during the 78 hour test directly after the Milena gasifier. The data given is corrected after recalibration of the different analysers.

Table III: Gas composition

	Duration test	Normal range
CO (vol% dry)	28.8	24 - 43
H_2 (vol% dry)	24.9	16 - 26
CO_2 (vol% dry)	20.4	10 - 21
CH_4 (vol% dry)	11.7	11 - 15
Ar (vol% dry)	0.1	0 - 1
C_2H_2 (vol% dry)	0.5	0.2 - 0.6
C_2H_4 (vol% dry)	4.2	3.7 - 5
C_2H_6 (vol% dry)	0.2	0.2 - 0.9
C_6H_6 (vol% dry)	1.1	0.8 - 1.3
C_7H_8 (vol% dry)	0.1	0.08 - 0.16
N_2 (vol% dry)	6.2	0.4 - 7
NH_3 (Vppm dry)	1485	-
HCl (Vppm dry)	3.2	-
HCN (Vppm dry)	88.9	-
H_2S (Vppm dry)	90.3	30 - 90
COS (Vppm dry)	3.9	4 - 9
Tar (g/m_n^3)	27.8	24 - 43
H_2O (vol% wet)	39	25 - 43
Gasifier temp. (°C)	851	750 – 875
Steam to biomass ratio (-)	0.28	0.02 – 0.35

The right column shows the range of measured concentrations when wood was used as fuel and olivine and sand as bed material.

4 CONCLUSIONS

The CO shift equilibrium is normally not reached in the Milena lab-scale gasifier. The hydrogen concentration is below theoretical values. Bed material selection can be used to influence the CO shift equilibrium. Olivine can be used as bed material if high hydrogen content is desired.

Methane yield is affected by temperature and bed material selection. A high methane yield is in favour of high overall efficiency to SNG. Operating the gasifier with sand at low temperature and a low steam to biomass ratio seems optimal, but gas cleaning equipment and catalysts downstream may set boundary conditions which result in different operating conditions.

Long term continuous operation (78 hours) of the Milena lab-scale gasifier coupled to the ECN gas cleaning infrastructure (Olga) is possible. Agglomeration of the bed material is not a problem when beech wood is used as a fuel.

The experiences gained from the different lab-scale tests have helped to design a Milena pilot plant of 0.8 MW_{th}. The pilot plant will be taken into operation in 2008. The pilot plant will be connected to the present pilot scale Olga gas cleaning test rig and will be used to demonstrate the production of SNG.

5 REFERENCES

[1] E.P. Deurwaarder, H. Boerrigter, H. Mozaffarian, L.P.L.M Rabou and A. van der Drift, Publication ECN-RX--05-194, available at www.ecn.nl.
[2] H. Boerrigter, S.V.B. van Paasen, P.C.A. Bergman, J.W. Köneman, R. Emmen and A. Wijnands, Report ECN-C--05-009, available at www.olgatechnology.com.
[3] S.V.B. van Paasen, J.H.A. Kiel, Publication ECN-C--04-013, available at www.ecn.nl.
[4] R. Rauch, K. Bosch, H. Hofbauer, D. Świerczyński, C. Courson, A. Kiennemann, Comparison of different olivines from biomass steam gasification.
5] R. Rauch, H. Hofbauer, I. Siefert, C. Aichernig, H. Tremmel, K. Voiglaender, R. Koch, R. Lehner, Steam gasification of biomass at CHP plant Guessing – Status of the demonstration plant.