PRESSURISED FLUIDISED BED GASIFICATION EXPERIMENTS OF BIOMASS AND FOSSIL FUELS

by

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Gasification of biomass and older fossil fuels, like brown coal, hot gas cleanup using a ceramic filter and combustion of LCV product gas in a combustor was performed using a 1.5 MW_{th} test rig (PFBG) at Delft University and a 10–50 kW_{th} at Stuttgart University (DWSA) in the framework of experimental pilot plant research on efficient, environmentally acceptable large scale power generation systems based on fluidised bed gasification technology.

The influence of operating conditions (pressure, temperature, stoichiometric ratio) on gasification performance (gas composition, conversion grades) was studied. The gasifiers were operated at pressures in a range of 0.15–1.0 MPa and maximum temperatures of circa 900 °C. The Delft gasifier has a 2 m high bed zone (diameter of 0.4 m) followed by a freeboard approximately 4 m high (diameter of 0.5 m). The IVD gasifier has a diameter of 0.1 m and has a total reactor length of 4 m. Both gasifiers are equipped with a hot gas cleanup ceramic filter and a pressurised combustor.

Measurements are compared with a model based on homogeneous elementary reaction chemistry and heterogeneous gas-char reactions related to emission of environmentally harmful components like fuel-nitrogen derived species. Results obtained are presented and analysed.

Carbon conversions were well above 80 %. Fuel-nitrogen conversion to ammonia is above ca. 50% and the highest for biomass in comparison to solid fossil fuel. The results are in-line with other pressurised fluidised bed gasification investigations with bottom feeding of biomass. Significant deviation with top feeding occurs. Measurements and model were in quite good agreement with each other.

Introduction

Power and heat production by pressurised air blown fluidised bed gasification of solid fuels is a promising technology regarding emissions and efficiency. These

process characteristics depend strongly on parameters like fuel type, pressure and air stoichiometry.

In the framework of co-operation between the partner institutes IVD (University of Stuttgart) and the section Thermal Power Engineering (Technical University Delft, TUD) TUD carried out experimental work at IVD. These experiments were performed with the advanced staged combustion (DWSA) installation. The thermal capacity of this test rig is *ca*. 50 kW(max). Also, measurements were done using the Delft pressurised fluidised bed (PFBG) test rig with a maximum thermal capacity of *ca*. 1.5 MW.

As fuels pelletised Miscanthus Giganteus (PFBG experiments) and German Brown Coal, from the Hambach open mine, and wood (DWSA test rig) have been selected. Brown coal is fuel on which a significant part of the electricity generation capacity in Germany (*ca*. 28%) and also in some Eastern European Countries, as well as *e. g.* Australia is based. Biomass has been selected as a fuel, because it is becoming increasingly relevant for *e. g.* the Dutch energy supply situation. The Dutch government has decided that by the year 2020 10% of the energy supply should be based on renewable sources, of which the main part will be biomass, see *e. g.* [1]. With energy supplied from biomass as a renewable source, there is almost no net CO_2 emission, as the CO_2 released to the atmosphere will be taken up by plants in a relatively short time scale. By substitution of coal or other solid fossil fuels by biomass, net CO_2 emissions will be reduced to a significant extend.

The research work described in this paper was primarily directed toward emission studies and the main goals were:
(1) to investigate the behaviour of the fuel conversion to main LCV gas species, and
(2) to study the fuel-Nitrogen conversion to NH_3 and HCN, which is directly linked to NO_x formation in combustion.

Other aspects of pressurised fluidised bed gasification were also studied, like tar composition in the generated low calorific value (LCV) gas and the carbon conversion. The hot gas filtration of the LCV gas was also of interest. Besides the fuel, the main gasification process parameters are the applied air stoichiometry and operating pressure.

Test facilities and main experimental variables

Figure 1 shows the pressurised fluidised bed test installation (DWSA) at IVD, University of Stuttgart. The DWSA has been used so far for several years in the framework of research in the field of conversion of solid fossil fuels under well-defined reproductive process conditions and on a small scale (50 kW_{th} maximal), see *e. g.* 2 . Figure 2 presents the PFBG test installation at Delft University.

Table 1 indicates the range within which the main DWSA and PFBG process variables can be varied. In Table 2 the main construction dimensions of both gasifiers are shown.

The DWSA installation can be divided into two main parts. The first part consists of an air preheater, fluidised bed reactor, solid fuel dosing vessel with on-line mass determina-
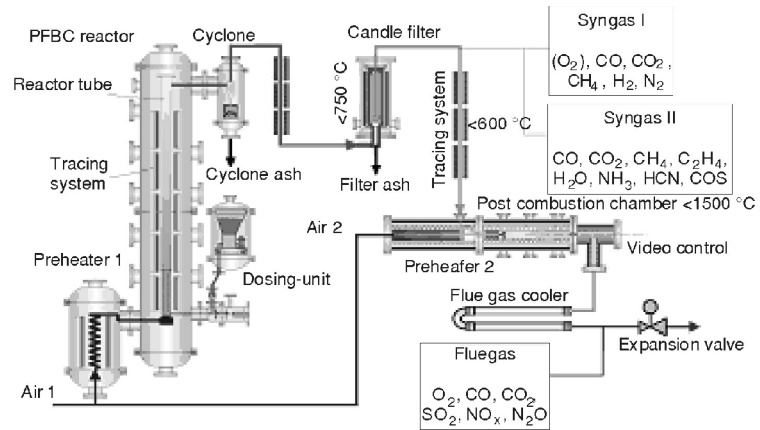
**Figure 1.
Schematic of the
DWSA test rig**

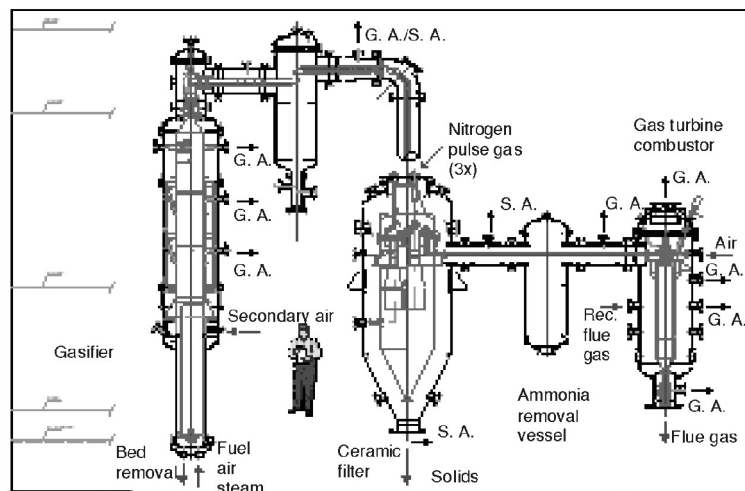**Figure 2.
Schematic of the
PFBG test rig**

**Table 1. Operating range (gasification) of the DWSA and the
PFBG test rig**

Variable	Range DWSA	Range PFBG
Pressure MPa	0.12–1.6	0.3–0.8
Temperature °C	750–1000	750–1000
Air stoichiometry, λ –	0.3–1.0	0.3–1.0
Fluidisation velocity m/s	0.1–1.0	0.5–0.8
Fuel	Coal, Brown coal, Biomass	Coal, Brown coal, Biomass

Table 2. Main dimensions of the DWSA and PFGB gasifer

	DWSA	PFBG
Bed diameter m	0.10	0.38
Max. bed height m	1.0	2.0
Freeboard diameter m	0.177	0.485
Freeboard height m	3.0	4.5

tion system and a hot gas cleaning section, containing a cyclone and a ceramic candle filter (Schumacher type). In the fluidised bed reactor the solid fuel is gasified with air to produce a low calorific value (LCV) gas that is cleaned of fly ash and unreacted solid carbonaceous material. Air and also additional Nitrogen can be preheated and is introduced into the reactor by four nozzles just above the distributor plate. The reactor is electrically heated in order to maintain a constant temperature over the bed as well as the freeboard section. The solid fuel is fed into the bed section in the bottom part just above the distributor by a screw feeder from beside the gasifier. The hot gas cleaning section ensures excellent gas-solid separation efficiency, with filter temperatures of about 500 °C.

The second part of the test rig consists of a combustion air preheater, a specially designed LCV gas burner, a flue gas cooler and a pressure control valve. The LCV gas combustor is situated in a water-cooled pressure vessel, and in the outer annulus secondary air is added.

The analysis of the produced LCV gas is performed directly behind the ceramic candle filter by means of continuous on-line O_2 (paramagnetic), CO- and CO_2 (NDIR) analysers. In addition, H_2, CO, CH_4 and N_2 concentrations are measured off-line by means of a gas chromatograph. An FTIR is used for measurement of NH_3, HCN, N_2O, NO, NO_2, CO, CO_2, CH_4, C_2H_4, C_2H_2, HCl, COS, and H_2O. Analysis of the combustion exhaust gases directly behind the combustor is performed by means of continuous on-line analysis for O_2, CO, CO_2, NO_x, N_2O, and SO_2. Besides gas analysis, also characterisation of solids has been performed. Samples of bed material, cyclone and filter ash have been taken. Proximate and ultimate analyses have been carried out as well as heating value determination.

The test rig at Delft University is an air/steam blown Pressurised Bubbling Fluidised Bed Gasifier (PFBG) with a ceramic channel-type filter and a modified pressurised AGT Typhoon gas turbine combustor for the Low Calorific Value (LCV) gas produced. Compressed air and pressurised steam enter the gasifier through a central nozzle in the distributor plate after preheating between inner and outer vessel. Fuel, bed material and additive are fed from big bags on a conveyor belt and transported into a double valve lock hopper system followed by screw feeding into a vessel. From there, the material is fed pneumatically into the bed through a feed point in the bottom plate and directed toward the central nozzle. Bed contents can be kept constant by a removal system at the bottom of the reactor. The freeboard is well insulated and only contains probes. The ceramic filter consists of three honeycomb-like elements that are cleaned on-line one at a time using pulses of heated Nitrogen. The combustor is of AGT Typhoon type and uses preheated air (350 °C) to combust the hot LCV gas.

Gas sampling with respect to the PFBG installation is done after the ceramic filter unit, before and after the combustor. An off-line operated FTIR with a heated 2 m gas cell is used to measure NH_3, HCN, N_2O, NO, NO_2, CO, CO_2, CH_4, C_2H_4, C_2H_2, HCl, COS, and H_2O. A GC is used to measure C_1–C_5 aliphatic hydrocarbons, Ar, CO_2, CO, and N_2 off-line. CO, CO_2, and SO_2 are measured on-line using NDIR analysers. A micro-GC is applied for on-line H_2 analysis. Paramagnetism based analysers are used for on-line O_2 measurement. An NDUV analyser is used for NO_x. A novel tar sampling method has been used, developed by [3]. Samples have been analysed by GC. Solids are sampled isokinetically before and after the ceramic filter unit.

The experiments concerning the DWSA test rig, reported in this paper, were performed with brown coal (Hambach open mine) and crushed pelletised wood sawdust. The experiments with the PFBG described in this paper were carried out with pelletised Miscanthus Giganteus.

Table 3 presents the main characteristics of the fuels. As can be seen, the wood species contains very low amounts of Nitrogen and Sulphur. It is also observed that the heating value of brown coal is somewhat higher. Also, the ratio fixed carbon: volatiles is different for the fuels involved, so that a different behaviour of the fuels is expected during gasification.

Table 3. Fuel analyses (Raw basis)

	Brown coal (Hambach)	Crushed wood pellets (Labee-A)	Miscanthus giganteus (Pellets)
Proximate analysis:			
Fixed carbon mass %	36.0	14.1	16.6
Volatiles mass %	44.1	76.7	71.5
Moisture mass %	15.6	9.2	9.1
Ash mass %	4.3	0.03	2.8
Ultimate analysis:			
C mass %	53.9	46.2	42.8
O mass %	36.0	47.4	47.2
H mass %	4.8	6.2	6.3
N mass %	0.6	0.04	0.5
S mass %	0.4	0.08	0.2
Cl mass %	0.1	0.0	0.2
Higher heating value:			
HHV MJ/kg	20.9	18.6	17.4

Experimental results

The DWSA experiments

Six experiments were performed using the DWSA installation. The average length of stable operation was between 3.5 to 4.5 hours. Almost 1.5 hours were necessary to reach steady state operation conditions.

Table 4 gives an overview of the main experimental results obtained using the DWSA test rig, regarding process conditions, LCV gas composition, heating value, carbon conversion and cold gas efficiencies. The main variables are the fuels applied, brown coal (BC) as a fossil fuel and crushed wood pellets (PW) as biomass species. For these fuels the air-stoichiometry was the most important process variable. With the wood experiments also pressure was varied.

Table 4. Overview of the DWSA test

Experiment	991103	991115	991118	991202	991206	991208
Pressure, P MPa	0.51	0.51	0.51	0.51	0.51	0.15
Bed temperature, T_b °C	791	802	858	782	824	792
Fuel	BC	BC	BC	PW	PW	PW
Fuel flow raw kg/h	3.7	2.5	2.2	3.3	3.0	1.6
Primary air flow kg/h	7.3	8.4	9.2	5.8	9.0	4.6
Primary air stoichiometry, λ –	0.30	0.51	0.66	0.30	0.48	0.51
Total air stoichiometry, λ_{tot} –	1.30	1.31	1.47	1.29	1.29	1.46
Nitrogen flow to gasifier, ϕ_{m,N_2} kg/h	2.5	2.8	2.7	7.2	3.0	1.4
LCV gas flow, $\phi_{m,LCV}$ kg/h	12.8	13.6	14.1	16.3	14.9	7.6
LCV gas composition CO vol%, wet	13.4	8.9	6.1	8.0	5.6	7.3
H_2 vol %, wet	10.4	7.0	4.5	5.2	3.8	5.3
CH_4 vol %, wet	2.1	1.4	0.6	3.0	2.0	2.1
C_2H_4 vol %, wet	0.1	0.1	0.1	0.5	0.3	0.7
CO_2 vol %, wet	10.9	11.8	12.6	9.7	13.7	13.3
H_2O vol %, wet	5.2	5.3	6.4	8.2	11.4	10.6
N_2 vol %, wet	57.3	64.8	69.0	65.1	62.7	60.2
Ar* vol %, wet	0.5	0.5	0.6	0.3	0.5	0.5
NH_3 vol %, wet	2290	1339	770	176	149	176
Higher heating value, HHV MJ/Nm³	3.91	2.65	1.65	3.21	2.20	3.21
Carbon conversion % (solid catch basis)	94	98	99	100	100	100
Cold gas efficiency	55.5	56.0	39.6	69.7	46.9	54.2
Fuel-N conversion to NH_3 %	70	60	41	100	100	100
Fuel-N conversion to char-N %	5	2	1	0	0	0

The LCV gas produced from the wood experiments was somewhat lower in calorific value as that from the brown coal experiments at practically the same pressure and air-stoichiometric values and bed temperatures. Compared on a purge N_2 free basis, the water concentration in the LCV gas from wood gasification was higher, whereas the H_2 concentration was lower, although there was less moisture and more elementary H in the wood as compared to brown coal. The light hydrocarbon concentrations were higher in the wood based LCV gas, whereas the CO concentration was significantly lower under the same conditions. The differences in the LCV gas composition are attributed to the structure in which C, H and O are bound in the fuels. Biomass consists of cellulose, hemi-cellulose and lignin. In older fossil fuels like brown coal, however, more aromatic molecular structures (pyridinic, pyrrolic) are present. This leads to different flash pyrolysis behaviour, the initial step together with drying in the fluidised bed gasification process. A different yield of initial products results from this dissimilar behaviour.

Carbon conversions measured were quite good and significantly above 90 % . Practically complete conversion was observed for biomass, which can be attributed to its higher reactivity as compared to brown coal.

In the wood gasification experiments, the fuel-Nitrogen was almost completely converted into NH_3, with HCN and solid bound Nitrogen being negligible considering experimental errors. The fuel-Nitrogen conversion into NH_3 with the brown coal experiments was lower compared to wood gasification. This has been reported in the literature before for a bottom-fed PFB, see *e. g.* [4]. The background of this different fuel- Nitrogen release behaviour can be explained to a certain extent by the different nature of the chemical bonding of Nitrogen, see *e. g.* [5] and [6]. In biomass, Nitrogen is mainly present in the form of peptide bounds (in *e. g.* amino-acids and proteïns).

In older fuels, like brown coal and (sub)bituminous coal, the Nitrogen species is more abundant in pyridinic and pyrrolic structures. There are also investigations which show different, lower conversions of fuel-Nitrogen into NH_3 with biomass, see *e. g.* [7]. The understanding of the devolatilisation mechanisms, however, is still not complete.

Differences can possibly also be attributed to the environment in which the primary fast pyrolysis takes place, either oxygen rich or lean.

The PFBG experiments

Gasification results

The fuel used for the PFBG gasification experiments was pelletised Miscanthus. Dolomite was added to the fuel in a mass ratio of *ca*. 0.03–0.05. Steam to air ratio was 0.04–0.11 in these tests.

Figure 3 shows an increase of the concentrations of light hydrocarbons, methane and ethylene, with decreasing air stoichiometry. The values are reasonably in line with the DWSA experiments, presented in table IV for wood.

Figure 4 shows an increasing trend of the higher heating value of the produced gas with decreasing air stoichiometry. The trend is in-line with earlier research concern-
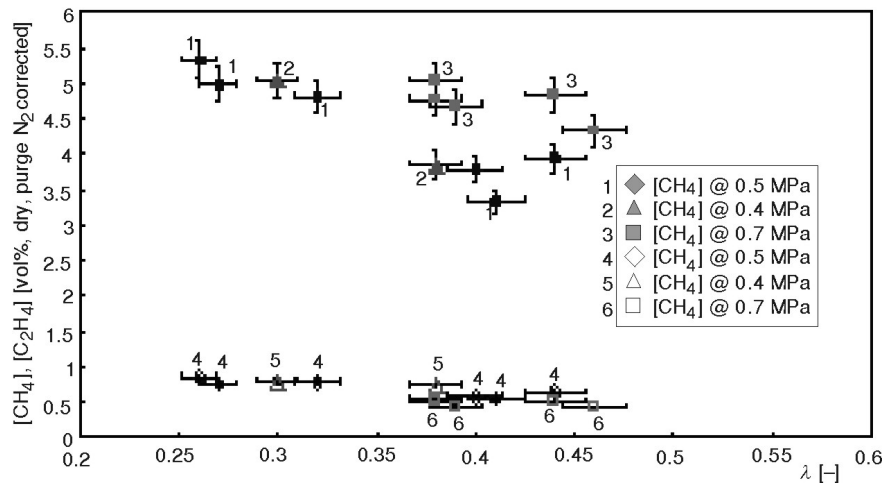
Figure 3. Light hydrocarbon species concentration versus air stoichiometry for the PFBG experiments
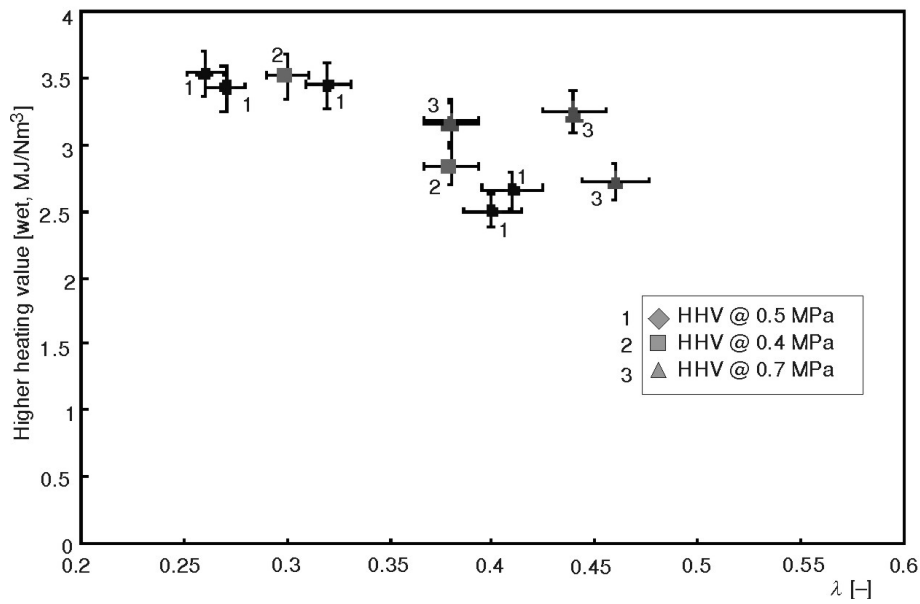

Figure 4. Higher heating value of the PFBG gasification product gas versus air stoichiometry

ing Miscanthus and Miscanthus/coal gasification experiments performed with the PFBG test rig, see *e. g.* 8 . The gas quality with respect to its higher heating value was sufficient for stable pressurised combustion in the downstream ALSTOM Typhoon gas turbine combustor, see 9 .

One of the major problematic groups of components in gas produced by biomass gasification is tar. They contribute to fouling of equipment (*e. g.* gas engine or turbines) and to emissions in gas cleaning and/or combustion processes. These components have been defined to be organic aromatic species with a molecular weight higher than benzene, see *e. g.* 10 .

Figure 5 shows an increase of the specific tar concentrations of Polyaromatic Hydrocarbons (PAH) and Phenols in the produced LCV gas. These compounds have been quantified by means of the novel solid phase adsorption technique, developed at KTH Sweden, see 3 . Especially the contribution of Phenols appears to be important at
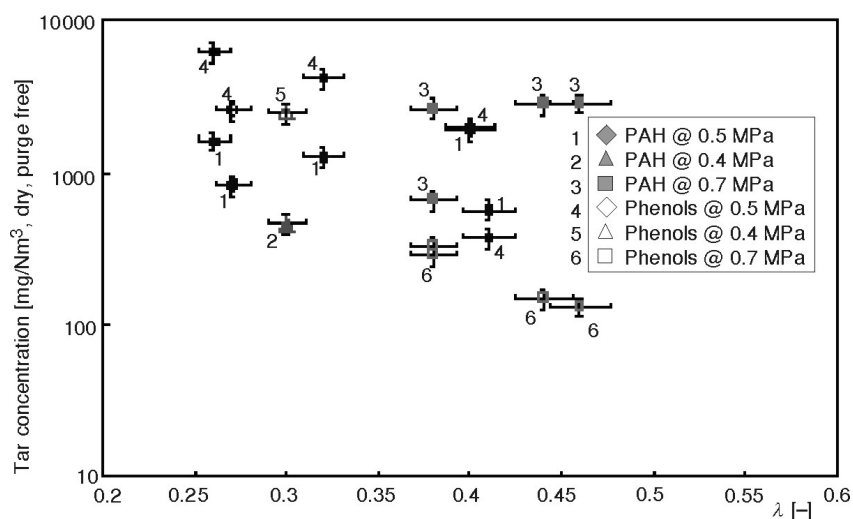

Figure 5. Higher hydrocarbon concentration versus air stoichiometry of the gasification product gas of the PFBG

lower Air Factors, accompanied with temperatures lower than 800 °C. For (pressurised) gasification using steam as (co-)gasifying medium the contribution of Phenols is also reported to be significant by 11 . Reproducible measurement of Benzene, Toluene and Xylenes with the s.p.a. sampling technique did not appear to be possible. The PAH's analysed range from Indene, Naphtalene to Pyrene, with Naphtalene being the major species.
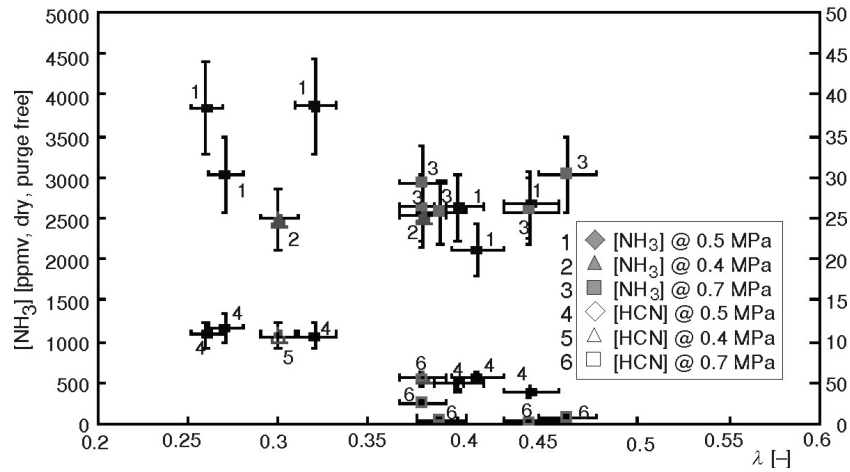
Figure 6. NO$_x$ precursor concentration versus air-stoichiometry in LCV gas produced by the PFBG gasifier

Figures 6 and 7 show the experimental PFBG results with respect to fuel-bound Nitrogen to Ammonia and Hydrogen Cyanide. These species are known precursors for NO$_x$ formation under *e. g.* gas turbine combustion conditions, which is a problem when dry, high temperature gas cleaning is applied, see *e. g.* 9 .
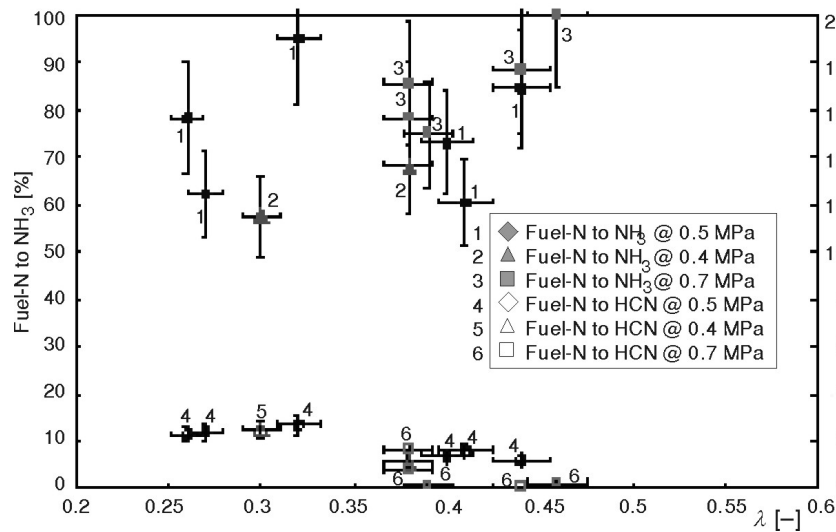

Figure 7. Fuel-Nitrogen conversion to NH$_3$ and HCN for the PFBG gasifier

From the results it can be concluded that a major part of the fuel-bound Nitrogen is converted to Ammonia. This has also been indicated before, in the description of the pressurised fluidised bed wood gasification using the DWSA test rig, although somewhat lower conversion values are observed in the PFBG tests.

The fuel-bound Nitrogen to NH_3 conversion values are comparable to and in-line with values reported by VTT, where a slightly smaller scale pressurised fluidised bed is operated (*ca.* 500 kW_{th}), for experiments with straw, a fuel quite comparable to Miscanthus, see *e. g.* 12 . During gasification of straw with dolomite as additive, these authors observed values in the range of 60–71 % fuel-Nitrogen conversion to NH_3, at air stoichiometry values between 0.28 and 0.31.

Significantly lower values of fuel-Nitrogen conversion to NH_3 were observed by researchers of KTH, where biomass (and coal) was gasified in a pressurised top-fed fluidised bed reactor, see 7 . Also, at KTH experiments were performed in a small scale atmospheric bubbling fluidised bed and a difference in top feeding as compared to bottom feeding was observed, see 13 . Top feeding led to significantly lower fuel-nitrogen conversion to NH_3. Differences were attributed to the environment in which the initial flash pyrolysis of the fuel takes place, *i. e.* reducing for top feeding versus oxidizing for bottom feeding.

Ceramic filter hot gas cleaning results

The ceramic filter unit in the Delft PFBG test rig has been operated for more than 100 hours under gasification conditions. They had to be exchanged once in the period for which the data presented here have been obtained. This was necessary, as due to opening of the unit to exchange a probe severe air leakage caused filter fire and cracking. All filter elements of the second set have been equipped with thermocouples then. The filters have been operated during long stable period set points, of which Figure 8 shows an example. Stable base-line pressure drop is observed, with values of the filter pressure drop between 10 and 16 mbar. Filter temperatures during stable set points (two periods in this
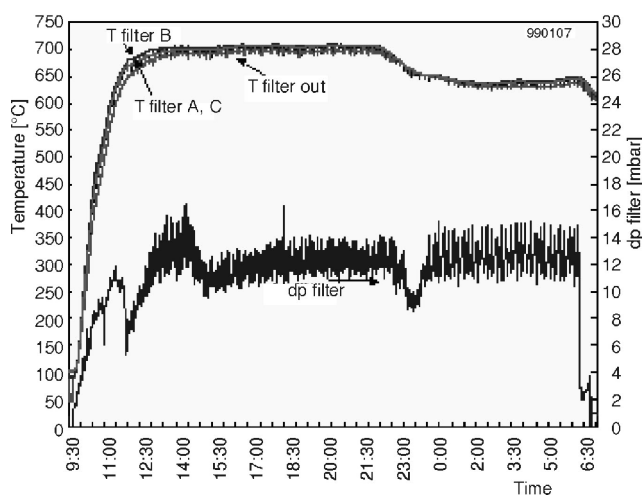

Figure 8. Characteristic temperatures and pressure drop behaviour cerning hot gas filtration for the PFBG test rig

case) of 650–700 °C are typical for all experiments carried out. The figure shows a transition from 0.7 MPa to 0.4 MPa, characteristic for a change of load in gasification (the air stoichiometry of the gasifier was kept constant for both set points).

Gas cleaning efficiencies of *ca*. 99.95% have been obtained, with typical filter outlet dust loads of 5–10 mg/Nm^3 LCV gas. These values are acceptable for gas turbine (combustor) operation considering also the sub-micron particle size of solids permeating the filter. The values are also well below Dutch emission standards for power producing stations as well as waste incinerators, see *e. g.* 14 .

Conclusions and future activities

The gasifiers at small scale (IVD) and PDU scale (Delft) were operated successfully regarding pressurised gasification and hot gas filtration. Carbon conversion and main gas composition were similar to literature data by VTT of their 500 kW$_{th}$ pressurised fluidised bed gasifier. Fuel-Nitrogen was converted mainly to NH$_3$. For biomass, the conversions were significantly higher than for brown coal, which is also in-line with VTT findings. A clear understanding of the Nitrogen release behaviour is still not complete, as differences exist in literature regarding different gasifier set-ups, which can be attributed to different devolatilisation behaviour of the solid fuels.

The gas cleaning by hot gas ceramic filtration was excellent, stable combustion of the LCV gas was ensured without significant fouling of the combustors.

Further PFBG research is necessary to extensively determine the influence of the fuel type used on Carbon conversion and the fate of tars, trace metals and NO$_x$ precursors. Also, gasification modeling is to be further optimised and validated with these and more experimental data to describe the processes of drying, devolatilisation, partial combustion and gasification.

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