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10th International Conference on Circulating Fluidized Beds and Fluidization Technology - CFB-10

Refereed Proceedings

Spring 5-2-2011

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Veronika Wilk
Bioenergy2020+ GmbH, veronika.wilk@bioenergy2020.eu

C. Aichernig
Repotec Umwelttechnik GmbH, Austria

H. Hofbauer
Repotec Umwelttechnik GmbH

WASTE WOOD GASIFICATION: DISTRIBUTION OF NITROGEN, SULPHUR AND CHLORINE IN A DUAL FLUIDISED BED STEAM GASIFIER

V. Wilk[1], C. Aichernig[3] and H. Hofbauer[2]
[1] Bioenergy2020+ GmbH, Wienerstraße 49, 7540 Güssing, Austria, veronika.wilk@bioenergy2020.eu, + 43 1 58801 17313
[2] Vienna University of Technology, Institute of Chemical Engineering, Austria
[3] Repotec Umwelttechnik GmbH, Austria

Abstract

Waste wood was gasified in a dual fluidised bed gasifier in order to investigate the behaviour of waste fuels in this technology. The distribution of nitrogen, sulphur and chlorine between the gasifier and combustor of the dual bed system was studied to identify the requirements for gas cleaning devices. The gasification system is suitable for the use of waste wood. A slight adaption of the gas cleaning equipment was necessary compared to gasification of natural woody biomass.

INTRODUCTION

In order to address the challenge of climate change and global warming the European Union agreed on the binding target to supply 20% of energy from renewable sources by 2020 (1). Solid biomass is an important renewable energy carrier, and it is going to play a major role in the future.

Steam gasification converts solid biomass into a high quality producer gas. Electricity and heat can be produced in an efficient way from this producer gas; it is also suitable for chemical synthesis of fuels and chemicals. At the Vienna University of Technology (VUT) the dual fluidised bed steam gasifier has been developed. The process was demonstrated in Guessing (Austria) in an 8 MWth gasification plant, that has operated successfully since 2002. In the meantime further plants are in operation or currently under construction. In all cases wood chips from natural sources are used as the feedstock.

The scope of this work is to investigate the suitability of waste wood for the gasification system described above. The use of waste wood in biomass gasifiers can increase the feedstock flexibility of the gasifier and could also offer some economic advantages. Furthermore, gasification is also an interesting approach for thermal waste wood treatment in general.

There are different types of waste wood according to precise quality standards. Slabs, logs or chippings, bark, fibreboards and surface treated wood are suitable for biomass combustion plants. Those plants use biomass from forestry as standard fuel. In Austria there are several power plants, where waste wood is combusted, among them the biomass plant in Timelkam and St. Veit an der Glan. The boilers are circulating fluidised beds (2), (3). However, contaminated wood, such as coal tar oil treated wood or wood-polymer-composites containing halogens, is not suitable for those plants and has to be treated in waste incineration plants.

Waste wood is already used in gasification processes at an industrial scale. In the autothermal gasifier in Lahti (Finland) waste wood is part of the feedstock mix (4). In the Amer power plant in the Netherlands 150.000 t/a of demolition wood are gasified. In both plants producer gas is combusted in a coal fired power station (5). The gasifiers are circulating fluidised beds with air as the gasification agent. Air gasification yields nitrogen-diluted producer gas with a low calorific value.

THE DUAL FLUIDISED BED GASIFIER

The dual fluidised bed gasifier is an allothermal gasifier, where gasification and combustion take place in spatially divided reactors. The principle is shown in Figure 1. The gasification reactor is fluidised with steam and a bubbling fluidised bed is formed. The residual ungasified char is transported into the combustion reactor together with the circulating bed material and is combusted with air. Combustion takes place in a highly expanded fast fluidised bed. Heat is delivered back to the gasifier by the bed material to satisfy the endothermic gasification reactions.

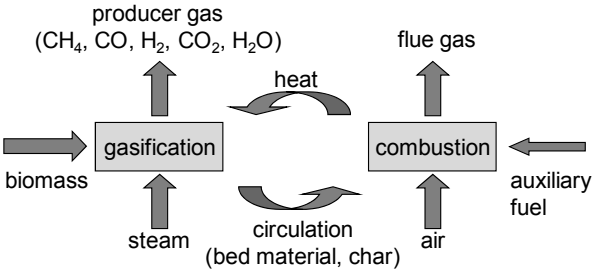

Figure 1: Principle of the dual fluidised bed gasifier

This gasification process has been demonstrated successfully in Guessing (Austria). Due to steam gasification the producer gas is virtually free of nitrogen. It is characterised by a high hydrogen content (>40%) and an average heating value of 12-14 MJ/m³ (stp) of dry gas. Producer gas is converted into electrical power in an internal combustion engine. Heat occurring in the process is fed into the local district heating system (6). This technology is about to be commercialised. Four new plants in the range of 10-20 MW are currently being built or are in the start-up phase.

The 100kW pilot plant

At the Vienna University of Technology a 100 kW pilot plant was installed. It is a prototype of the Guessing gasifier, and has been used for the design of the Guessing gasifier and is now used to further develop the dual fluidised bed gasifier. In Figure 2 the pilot plant is schematically illustrated.

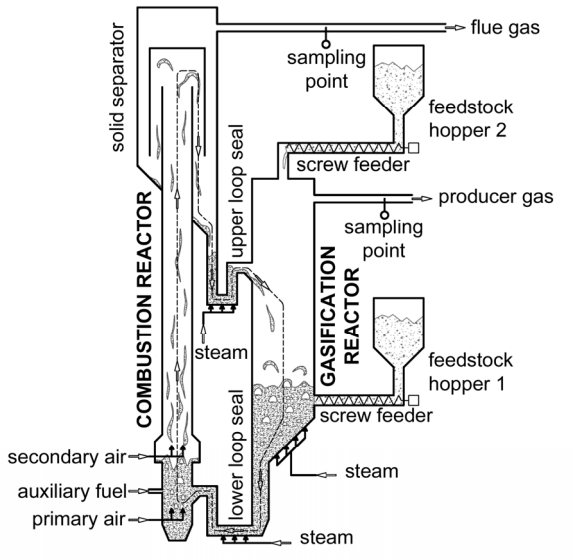

Figure 2: 100 kW dual fluidised bed pilot plant for steam gasification

Superheated steam for gasification is produced in an electrically heated steam generator. The gasification reactor and the combustion zone are connected by loop seals. In order to promote transport of solids and to prevent leakage the loop seals are also fluidised with steam. In the combustion reactor heat is generated as residual char is combusted with air. At the bottom of the combustor primary air is injected and a dense fluidised bed is formed in the bottom region. Light fuel oil is used as auxiliary fuel; it is also added there. The temperature in the gasifier is controlled by the amount of oil inserted. Usually the temperature difference between the gasification zone and the combustor is about 70 to 100°C depending mainly on the water content of the fuel and the circulation rate. Secondary air is injected at a higher level to transport particles to the top of the riser. Hot bed material is transported back into the gasification reactor. In this gasification process olivine is used as the bed material. It has been proven that olivine has good mechanical stability and shows moderate tar cracking activity (7).

After leaving the gasifier the producer gas is cooled in an oil-cooled heat exchanger to temperatures around 250°C and is sampled for gas analysis. Producer gas and flue gas are mixed and combusted in an afterburning chamber with air. A cyclone removes particles before passing to the stack.

Measurement equipment

During the experiments, producer gas and flue gas properties are measured. The main producer gas compounds are analysed by a Rosemount NGA2000 device. The range of measurement is 0 to 100% for CH_4, CO, H_2, and CO_2 and 0 to 25% for O_2. A gas chromatograph (Syntech Spectras GC 955) is employed for the online-measurement of N_2, C_2H_4, C_2H_6 and C_3H_8.

An impinger bottle method for tar measurement has been developed at VUT. It is similar to the conventional tar protocol, but has been adapted for producer gas from steam gasification. Toluene is employed as a tar absorbent. Dust, entrained char,

water and tar content can be analysed from one sample. Further description of the tar measurement is available in (8). NH_3, HCl and H_2S are also measured by the impinger bottle method. The ammonia content is determined by dissolution in H_2SO_4 with a molar concentration of 0.05. Ammonium sulphate is formed and detected by ion chromatography. The concentration of HCl is analysed by dissolution in H_2O_2 in impinger bottles. Chlorine is measured by ion chromatography. For the measurement of H_2S impinger bottles filled with 35%-KOH are used. H_2S is detected by potentiometric titration.

In the flue gas stream the CO, CO_2 and O_2 content is measured by a Rosemount NGA2000 device. The range of measurement is 0 to 100% for CO and CO_2 and 0 to 25% for O_2. For the assessment of NO and SO_2 a Rosemount NGA2000 MLT4 device is used, which allows online measurement. The HCl concentration in the flue gas stream is determined by impinger bottles filled with H_2O_2. Chlorine is measured by ion chromatography.

FEEDSTOCK CHARACTERISATION

Two different types of waste wood have been gasified in the pilot plant at VUT. Waste wood A has been provided by a manufacturer of windows and doors. It contains pieces of coated chipboards, fibreboards, surface treated wood and cardboard. It is in the form of chips with particle sizes in the range of 10 to 30 mm with a considerably high amount of fine particles. Waste wood B mainly consists of shredded furniture in the form of chips and fibres ranging from 10 to 40 mm. The content of fine particles is higher than in waste wood A. Table 1 gives an overview at the elementary analyses.

Table 1: Composition of wood pellets, waste wood A and waste wood B

		wood pellets	A	B
water content	%	6.11	6.73	15.49
volatile matter	%, dry	86.45	81.39	74.82
ash content	%, dry	0.29	1.56	7.90
carbon	%, dry	50.23	48.31	48.71
hydrogen	%, dry	6.04	5.51	4.78
oxygen	%, dry	43.67	43.64	36.41
nitrogen	%, dry	0.05	2.49	1.99
sulphur	%, dry	0.005	0.03	0.08
chlorine	%, dry	0.003	0.02	0.13
net calorific value	kJ/kg	18 753	18 420	17 719

Soft wood pellets are the standard feedstock for the pilot plant, as they are standardised fuel with defined water content and heating value. A significant ash content has been measured in waste wood B. Therefore, there is less volatile matter in this sample. The water content is also markedly higher in waste wood B. Soft wood pellets contain virtually no nitrogen, sulphur and chlorine. Noxious gases are formed from these elements. Waste wood A contains 2.49% of nitrogen, which is present in adhesives and coatings. The nitrogen content of waste wood B is in a comparable range. The chlorine and sulphur content in waste wood B exceeds the sample of waste wood A.

GASIFICATION EXPERIMENTS

During all experiments the main operating conditions of the gasifier are kept constant in order to achieve similar reaction conditions and comparable results. The fuel input is about 100 kW. The characteristic temperature in the gasifier is 850°C in all experiments. The steam-to-carbon ratio, which is determined by fluidisation settings and water content of the feedstock, is in the range of 1.8 to 1.9. For one experiment stable gasification of feedstock lasts an average for 6 to 8 h.

During gasification of waste wood B more light fuel oil was required to maintain the temperature in the gasification reactor. Due to the high water content more energy is consumed to evaporate the water in the feedstock, which results in a greater requirement of auxiliary fuel.

In Figure 3 the main producer gas compounds are illustrated. All experiments yield a similar gas composition, which is typical for steam gasification of woody biomass. Gasification of waste wood decreases the hydrogen content, but increases CO and C_2H_4 yield. Because of the higher nitrogen content in waste wood, a significant concentration of ammonia has been detected. H_2S and HCl content increases with increase of sulphur and chlorine in the feedstock.

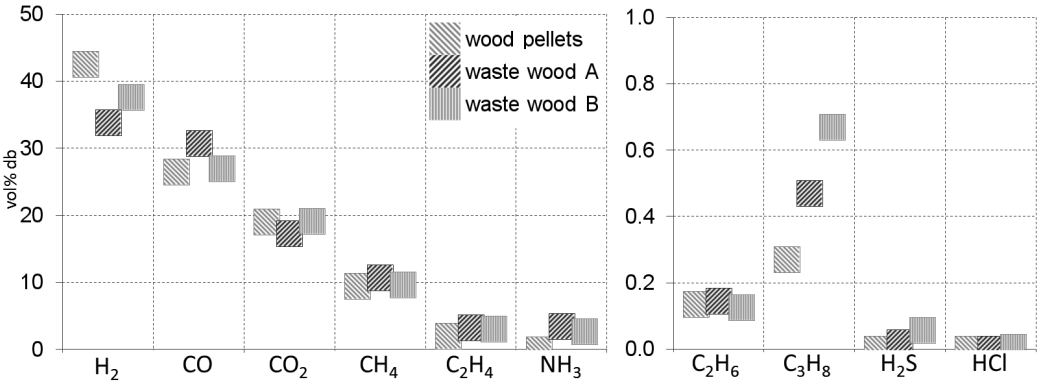

Figure 3: Producer gas composition in vol% referred to dry gas

Table 2: Solid impurities and tar in producer gas

		wood pellets	A	B
dust content	g/Nm³, db	8.3	18.8	>>
char content	g/Nm³, db	37.7	31.8	>>
GC/MS tars	g/Nm³, db	5.5	12.4	>>
gravimetric tars	g/Nm³, db	1.8	7.8	>>

Table 2 gives an overview of the solids and tars in the producer gas. Gravimetric tars are weighed after evaporation of the solvent. A GCMS device is used to measure the content of many different tar species. As lighter hydrocarbons (especially indene and naphthalene) disappear during evaporation and not all higher hydrocarbons are analysed by GC/MS, the measuring range overlaps. The content of tars and inorganic dust increases significantly, when waste wood is gasified. Waste wood B contains high amounts of fine particles and ash, which are found in

the producer gas. The particle content of producer gas from waste wood B is so high, that it is not possible to take a sample for tar measurement. Thus, there is no quantitative information on tar, dust and char for the time being.

DISTRIBUTION OF POLLUTANTS IN THE DUAL FLUIDISED BED GASIFIER

Due to the spatial separation of the gasification and combustion reactors, two gas streams, producer gas and flue gas, are generated. Volume flows of producer gas and flue gas are calculated with IPSEpro, an equation-oriented steady state simulation software. Due to measurements the mass and energy balances form an over-determinated equation system, which is solved by the Method of Least Squares. More details about this procedure can be found in (9). Flow rates of the equilibrated solution represent the basis for balancing trace elements like N, S and Cl.

Balance of nitrogen

Figure 4 shows the distribution of the main nitrogen compounds in the pilot plant during the gasification of waste wood. There is N_2 in air injected into the combustion zone and N_2 to flush the hopper. Output streams are NH_3 and N_2 in the producer gas, and N_2 and NO in the flue gas. In the illustration N_2 in combustion air, in the hopper flush and flue gas is omitted, because it does not react. The balance shows that the vast majority of nitrogen (ca. 90%) is present in the producer gas in the form of NH_3 (waste wood A = 3.4 vol%, waste wood B = 2.7 vol%). Only a small portion of fuel nitrogen is transported to the combustion section with the char. There it is oxidised, NO averages 190 ppm (waste wood A) and 110 ppm (waste wood B). During gasification also other nitrogen compounds such as HCN can be formed; they have not been analysed yet.

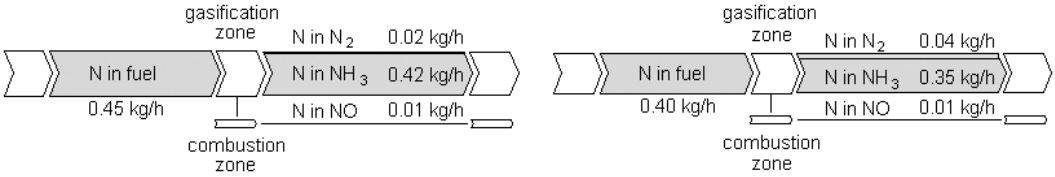

Figure 4: Distribution of nitrogen (left: waste wood A, right: waste wood B)

Balance of sulphur

As sketched in Figure 5, input streams containing sulphur are fuel and light fuel oil.

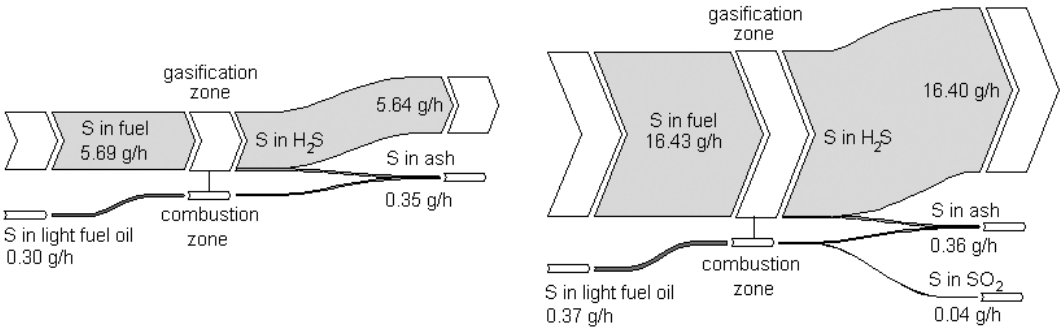

Figure 5: Distribution of sulphur (left: waste wood A, right: waste wood B)

In the producer gas concentrations of 210 ppm (waste wood A) and 570 ppm (waste wood B) were measured. A small amount of sulphur sticks to ash. In the pilot plant, producer gas and flue gas are mixed and combusted, then particles are separated in a cyclone. Thus, producer gas ash and flue gas ash are analysed as a mixture. When waste wood A is gasified, the SO_2 concentration in the flue gas is below the detection limit. Flue gas of waste wood B contains on average 5 ppm of SO_2. Other sulphur compounds that can be formed during gasification have not been analysed yet.

Balance of chlorine

Fuel is the only source of chlorine in the gasification process (Figure 6). Minor contents of HCl are measured in the producer gas, 35 ppm for waste wood A and 70 ppm for waste wood B. The majority of chlorine is bound to ash particles (more than 90%). HCl in the flue gas was below the detection limit in all experiments. Other chlorine compounds that might occur have not been determined yet.

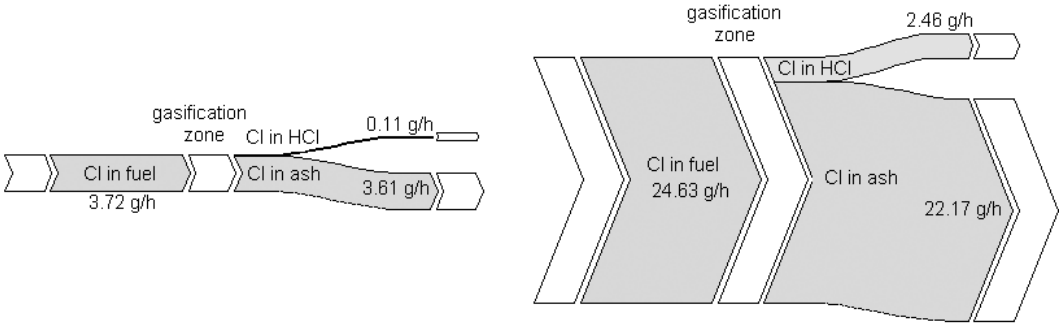

Figure 6: Distribution of chlorine (left: waste wood A, right: waste wood B)

GAS CLEANING

The distribution of pollutants in producer gas and flue gas is an important basis for adaption of the gas cleaning system for use of waste wood. The balance of pollutants shows that nitrogen and sulphur are mainly present in producer gas and chlorine is mainly found in ash.

At the demonstration plant in Guessing, producer gas cleaning consists of the following devices: a precoated bag house filter to remove particles and an organic scrubber to precipitate tars. That is sufficient for gasification of wood chips from the forest. If the system has to be adapted to waste wood, some changes will be necessary. The majority of chlorine is bound to particles, which are removed in the bag house filter. It has been shown that the precoat material, which is injected at the Güssing plant prior to the fabric filter, captures chlorine too (10). Thus, no additional effort might be necessary for chlorine. In the scrubber water in the producer gas is condensed. If the scrubber is operated at a low pH value, ammonia will be dissolved in water and will also be precipitated (11). Depending on the concentration of H_2S in the producer gas and legal requirements an additional scrubber for sulphur might be necessary. Only traces of pollutants are present in flue gas in the pilot plant. As a first assessment, no adaption of the flue gas cleaning equipment (consisting only of a bag house filter) is necessary.

CONCLUSION

Gasification experiments with two different types of waste wood have been carried out in the pilot plant at the Vienna University of Technology. From these experiments it can be concluded that the dual fluidised bed system is suitable for the gasification of waste wood. The majority of pollutants is present in the producer gas. Thus gas cleaning equipment in the producer gas line has to be adapted according to the higher concentration of pollutants in the waste fuels.

ACKNOWLEDGEMENT

We want to thank the team of the Notified Testing Laboratory for Combustion Systems at VUT for their support. Many thanks to EnergieAG Oberösterreich for providing the waste wood samples. Bioenergy2020+ is funded within the Austrian COMET program managed by the Austria Research Promoting Agency FFG. The financial support of the funding association FFG and the company partners Magna, Repotec and BKG is gratefully acknowledged.

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