In-Bed Catalytic Tar Reduction in a Dual Fluidized Bed Biomass Steam Gasifier

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A nickel-enriched catalytic bed material was tested for tar reduction in a 100 kW_{th} dual fluidized bed biomass steam gasifier. Gas composition and tar content were measured after the reactor and compared with data from gasification tests without a catalytic bed material. H_2, CO, CO_2, and CH_4 contents in the product gas, as well as tar conversion rates, are reported for different amounts of catalytic active bed material and different operating conditions. Water conversions, gas yields, and lower heating values were calculated. The catalyst showed no noticeable deactivation in two tests of 30 and 45 h. These results obtained at the pilot scale represent an important intermediate step in preparing the technical breakthrough of dual fluidized bed biomass steam gasification.

Introduction

Gasification of biomass is an attractive technology for combined heat and power production. Although a great deal of research and development work has been carried out during the past decade the commercial breakthrough for this technology is still far away. One problem that has not been completely solved so far is the tar content in the product gas, which can cause plugging in the colder parts of the plant.

Fluidized bed gasification is suitable for various biofuels and is not restricted to a small range of particle sizes and water contents as is the case for fixed bed gasifiers. Typical tar contents in product gas from fluidized bed gasifiers are between 2 and 20 g/Nm^3 of dry gas.[1] These contents are far too high to avoid the problems mentioned above if the gas is cooled to 50 °C, which is common for gas engines. Therefore, tar reduction before cooling is essential.

One method that has been studied intensively is catalytic tar destruction.[2−6] Recently, overviews of the current knowledge on the catalytic elimination of tars were published.[7,8] Catalytic materials such as dolomite, alkali metals, and nickel-based catalysts are under investigation. Either a catalytic bed can be located downstream after the gasifier, or the catalytic active material can be put directly into the gasifier itself. Corella et al.[9] made an excellent and careful comparison between the two possibilities and found no significant difference in their effectiveness concerning tar reduction. The effectiveness of dolomite in a second reactor was only slightly higher than the effectiveness of the in-bed location.

Use of an in-bed catalyst (as the bed material or as an additive to the feed) eliminates at least one reactor and therefore reduces investment costs. Attrition and deactivation of the catalyst due to carbon deposition at the catalyst have been reported by several research groups.[7] Of course, the catalyst can be regenerated by burning off the carbon layer. This can be achieved without the necessity of removing the catalyst from the reactor by using a dual fluidized bed design in which the bed material circulates from the gasification zone to a combustion zone where the regeneration of the catalyst takes place.[10] Therefore, no deactivation should be expected in the case of a dual fluidized bed gasifier. Some results of investigations with dual fluidized beds using a catalytic active bed material are presented in this paper.

Table 1. Results of the Fuel Analysis

parameter	value	units
C	49	mass % wf
H	6.52	mass % wf
N	0.12	mass % wf
S	<0.05	mass % wf
O	44.31	mass % wf
ash	0.26	mass % wf
LHV	17 120	kJ/kg
HHV	18 620	kJ/kg

Pilot Plant and Feedstock

Feedstock. Wood pellets with a biomass feed of 25 kg/h_{wet} were used as the fuel for all experiments. The water content of the wood pellets was 7.3 wt %. The water-free (wf) composition of the fuel is reported in Table 1.

A small amount of an additional fuel was used to control the temperature of the gasification zone. This additional fuel was light fuel oil (LHV_{add_fuel}, 42.700 kJ/kg), which was inserted by a squeezed tube pump.

Pilot Plant. The basic idea of the dual fluidized bed steam gasifier is to divide the fluidized bed into two zones, a gasification zone and a combustion zone. A circulation loop of bed material is created between these two zones, but the gases should remain separated. The circulating bed material acts as heat carrier from the combustion zone to the gasification zone. The basic principle of this process is illustrated in Figure 1.

Biomass is fed into the gasification zone and gasified with steam. The gas produced in this zone is therefore nearly free of nitrogen. The bed material, together with some charcoal, circulates to the combustion zone. This zone is fluidized with air, and the charcoal is burned. Additional fuel can be used to control the temperature, which is important for optimized operation of the gasifier. The exothermic reaction in the combustion zone

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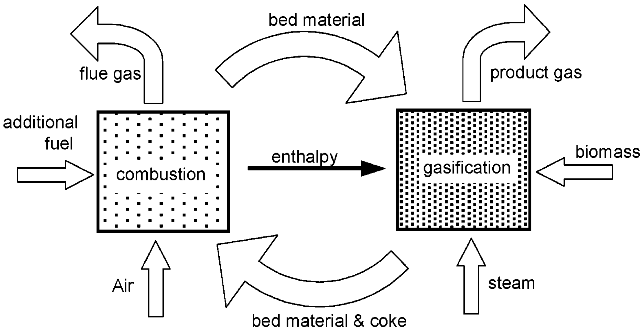

Figure 1. Principle of the dual fluidized bed steam gasifier.[10]

provides the energy for the endothermic gasification with steam. Therefore, the bed material has a higher temperature at the exit of the combustion zone than at the entrance. The flue gas is removed without coming into contact with the product gas. With this concept, it is possible to obtain a high-grade product gas without the use of pure oxygen. This process can be realized with two fluidized beds connected with transport lines or with an internally circulating fluidized bed as shown here.

Figure 2 shows a simplified flow sheet of the 100 kW_{th} pilot plant used in this study. Two screws are used to feed the biomass into the gasification zone. The first one is equipped with a frequency converter to control the amount of fuel. To keep the biomass in close contact with the bed material, the biomass is inserted by the second screw directly into the fluidized bed, which is important especially in case of catalytically active bed materials.

The main fan provides the necessary amount of air for the combustion zone. The air stream is divided into primary and secondary air flows, which are controlled separately. The secondary air is preheated (500 °C) electrically.

To reach the required gasification temperature, additional fuel (light fuel oil) is inserted into the combustion zone through a nozzle. To prevent thermal destruction of the oil, the nozzle is cooled by air. The amount of oil added depends on the temperature in the gasification zone.

During the gasification process, the siphon and the gasification zone are fluidized with steam. The siphon and the gasifier inlet pipes are covered with heating elements to superheat the steam needed as the gasification agent during the gasification process as well as the air during the start-up period.

The product gas leaves the gasification zone and is cooled to temperatures of about 150−350 °C with a heat exchanger, which is operated with a thermo oil at temperatures up to 300 °C. The heat of the thermo oil is extracted by a water heat exchanger. After cooling, the soot and fine abraded bed material are separated from the product gas either by a cyclone or by a precoated bag filter. The tar content of the dedusted gas is reduced by a scrubber. The scrubber liquid used is rape seed oil methyl ester, which shows excellent properties for this purpose.

To minimize the pressure difference between the gasification and combustion zones, a throttle valve is installed in the flue gas pipe, which is able to operate up to temperatures of 1000 °C. This valve is automatically controlled depending on the pressure drop over the product gas cleaning line.

The clean product gas is mixed with the flue gas and combustion air and is burned in a cyclone, which, in this case, is a separator for particles as well as a combustion chamber. To ensure complete combustion of the gas, a pilot burner is in continuous operation. The flue gas is cooled by dilution with ambient air (shown as a heat exchanger in Figure 2) and finally transported to the environment via a chimney.

Tar Sampling and Analysis. The measurement of dust content is performed according to standard ONORM M5861-1. The dust and tar contents are measured after the heat exchanger, after the bag filter and cyclone, and after the scrubber. The measurement of the tar is carried out similarly to the tar protocol “gravimetric tars”.[11] A small amount of product gas is sampled isokinetically for a certain period of time. Dust and heavy tars ($T_B > 200$ °C) are deposited in a filter cartridge filled with glass wool. Low-boiling tars ($T_B <$ 200 °C) are washed out with toluene using washing flasks, which are operated at a temperature of about −20 °C. Along with the tar content, the particle and moisture contents are also measured. A gas meter and a thermocouple are used to determine the dry product gas stream.

Samples of the toluene from the washing flasks are taken for the characterization of the tars using a gas-phase chromatograph coupled with a mass spectrometer. The tar composition at the gasifier exit depends

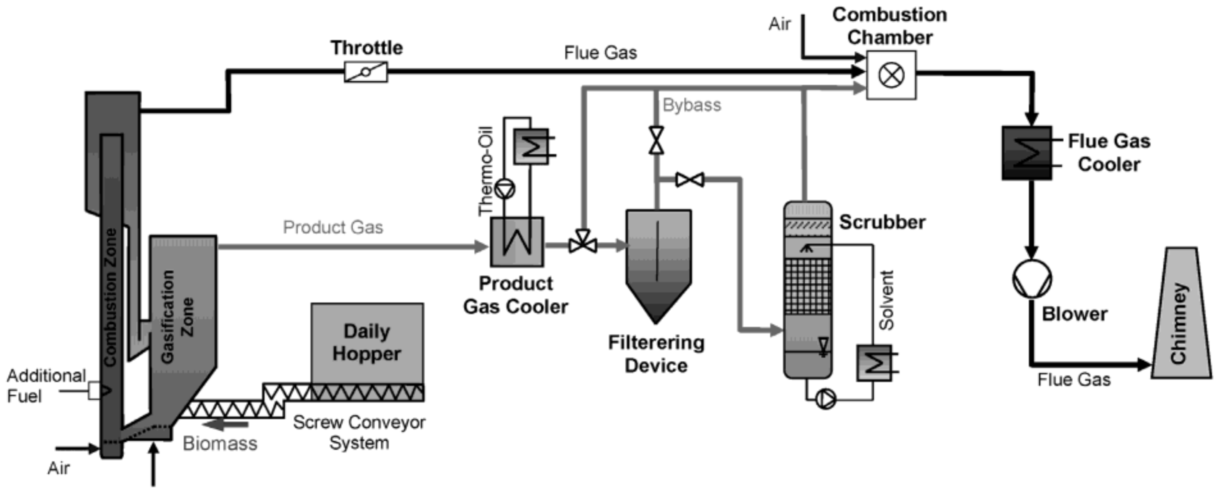

Figure 2. Simplified flow sheet of the pilot plant.

Table 2. Chemical Composition (wt %) of the Olivine[15] and Catalyst Used

component	olivine	catalyst
MgO	48.0−50.0	46.2−48.1
SiO_2	39.0−42.0	37.6−40.4
Fe_2O_3	8.0−10.5	7.7−10.1
$Al_2O_3 + Cr_2O_3 + Mg_3O_4$	0.8	0.8
CaO	<0.4	<0.4
NiO	<0.1	3.7

Table 3. Ranges of Parameters

parameter	values						
temperature (°C)	750	800	850[a]	900			
steam/fuel		0,3	0,6[a]	0,9			
catalyst/olivine			0[a]	5	10	20	43

[a] Reference value.

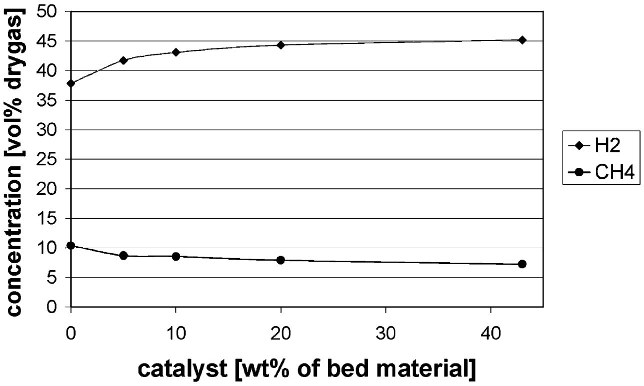

Figure 3. Hydrogen and methane concentrations for various amounts of catalyst (gasification temperature = 850 °C, steam-to-fuel ratio = 0.6).

on a number of operating parameters, as well as on the amount of catalyst in the bed material. Just as an example, a typical tar composition (850 °C, steam/fuel ratio of 0.6, 10 wt % catalyst) at the gasifier exit is 64 wt % naphthalene, 16 wt % acenaphthylene, 5 wt % acenaphthene, 8 wt % fluorene, 6 wt % phenanthrene, and 1 wt % indene.

The gas composition of the product gas was analyzed with an offline gas-phase chromatograph coupled with a thermal conductivity detector (tcd). The main components (H_2, N_2, CO, CO_2, CH_4, C_2H_6, and C_2H_4) were measured every 30 min. Gas compositions reported in this paper are averaged values with respect to time on stream.

The gas composition and tar content of the product gas were measured after the product gas heat exchanger.

Catalyst Used. The catalyst employed in this study is a Ni/olivine catalyst that was developed at the University of Strasbourg (ECPM, Strasbourg, France) and first applied to a fluidized bed biomass steam gasification bench process at the University of L'Aquila (L'Aquila, Italy). The development and production are described in several publications[5,12] and patents.[13,14] The catalyst is a modified olivine enriched with nickel using nickel nitrate. The catalyst contained 3.7 wt % of nickel on the olivine after calcination at 1100 °C. This calcination temperature results in nickel oxide that is strongly linked to olivine (grafted NiO) but is still reducible under gasification conditions. Strong metal−support interactions provide mechanical resistance to attrition, which is important in fluidized bed use, and resistance to coking is provided by the absence of Ni particle sintering.

The mean size and the size distribution of the catalyst are similar to those of natural olivine, which is usually used in the 100 kW pilot plant. Therefore, it can be stated that the fluid dynamic behavior of the catalyst is similar to that of natural olivine and, therefore, that the catalyst is mixed ideally with the natural olivine inside the fluidized bed. Different mixtures of olivine and catalyst were used during the experiments (catalyst comprising 0, 5, 10, 20, and 43 wt % of the bed material).

Olivine is actually a name for a series between fayalite and forsterite. Fayalite is the iron-rich member of the series with a pure formula of Fe_2SiO_4. Forsterite is the magnesium-rich member with a pure formula of Mg_2SiO_4. The two minerals form a series in which the iron and magnesium can be substituted for each other without a large effect on the crystal structure. Because of its iron content, fayalite has a higher index of refraction, is heavier, and has a darker color than forsterite. Otherwise, these materials are difficult to distinguish, and virtually all specimens of the two minerals contain iron and magnesium. For simplicity, they are often treated as one mineral, olivine. The chemical composition of the natural olivine sample used here is listed in Table 2. This olivine has a density of $3250-3300$ kg/m^3, a grain size of 400−600 μm, and a total porosity of 13−14%.[15]

Experimental Results

Range of Investigations. The experiments were carried out at different gasification temperatures (750, 800, 850, and 900 °C). The gasification temperature is defined as the temperature of the fluidized bed at the height of the biomass feeding point. Further parameters that were varied include the steam-to-fuel ratio (0.3, 0.6 and 0.9 $kg_{H_2O}/kg_{dry\ fuel}$) and the catalyst/olivine mixture (0-43 wt%). The steam-to-fuel ratio is calculated according to eq 1. As reference conditions, a temperature of

$$\mathrm{sfr} = \frac{\dot{m}_{\mathrm{w\_fuel\_in}} + \dot{m}_{\mathrm{w\_fluid\_in}}}{\dot{m}_{\mathrm{fuel\_dry\_in}}} \qquad (1)$$

850 °C and a steam-to-fuel ratio of 0.6 were defined on the basis of previous experience. Table 3 lists the ranges of variation of the three main parameters.

Gas Composition. It is well-known from the literature that, at temperatures between 750 and 900 °C, Ni catalysts in combination with biomass gasification processes decrease the tar content of the product gas and increase the hydrogen volumetric content.[16−19] Consequently, the methane content decreases as well as the CO_2 content.[20] To close the mass balance, the CO content has to increase, as is validated by the measurements.

In Figure 3, the dependency of the concentrations of hydrogen and methane in the dry gas on the amount of catalyst in the bed material is shown for the reference conditions. Use of the artificial Ni catalyst leads to an increased amount of hydrogen and a decreased amount of methane in the product gas. A higher steam-to-fuel ratio also results in a higher conversion rate, which leads to an enhanced hydrogen content in the dry product gas. The same tendencies can also be observed without addition of any catalyst, that is, with natural olivine.[21] It was generally observed that the addition of the catalyst intensifies the quantity of the effects but

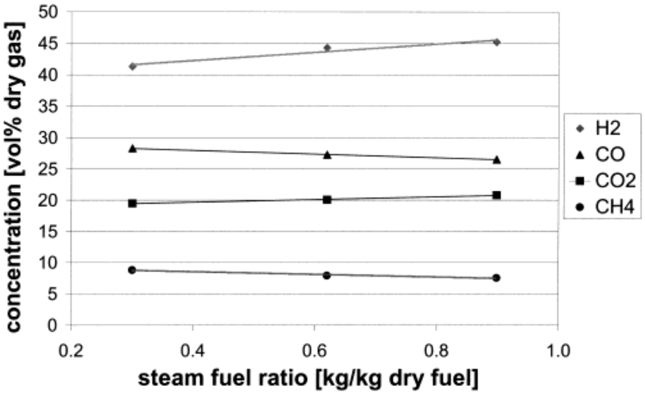

Figure 4. Dry gas composition as a function of steam/fuel ratio (gasification temperature = 850 °C, 20 wt % catalyst).

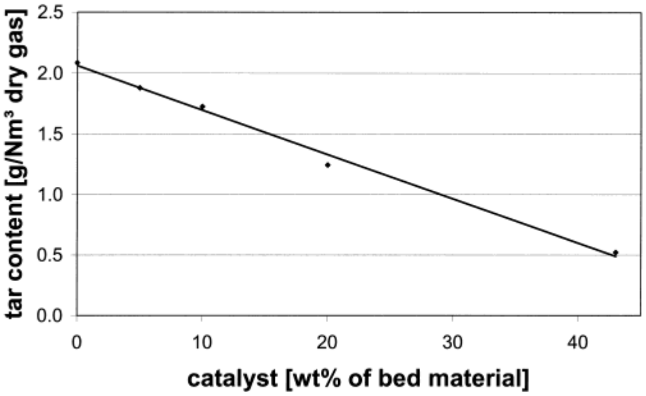

Figure 5. Tar content as a function of amount of catalyst.

does not change the tendencies. The same applies also for the correlation between the dry gas composition and the steam-to-fuel ratio. In Figure 4, this coherence is shownfor a gasification temperature of 850 °C and a catalyst content in the bed of 20 wt %. Within the investigated range of steam-to-fuel ratios (0.3−0.9), the methane and carbon monoxide contents decrease, whereas the hydrogen and carbon dioxide contents increase. These results were found independently of the different catalyst−olivine mixtures used (0−43 wt %).

Tar Content. The tar content in the product gas was measured after the heat exchanger at about 200 to 250 °C, and the values were averaged with respect to time on stream. In Figure 5, the influence of the amount of catalyst in the bed material on the tar content in the dry product gas is shown. It can be clearly observed that the tar content is decreased from more than 2 to 0.5 g/m_n^3 simply by the use of an in-bed catalyst.

These experiments were carried out at a gasification temperature of 850 °C and a steam-to-fuel ratio of 0.6. As expected, the tar content in the dry product gas decreases with increasing amounts of catalyst in the bed material. A reduction of more than 75% could be achieved using a catalyst content of 43 wt % of the bed material. This concentration is low enough for the product gas to be used in a high-temperature fuel cell without any additional tar removal.

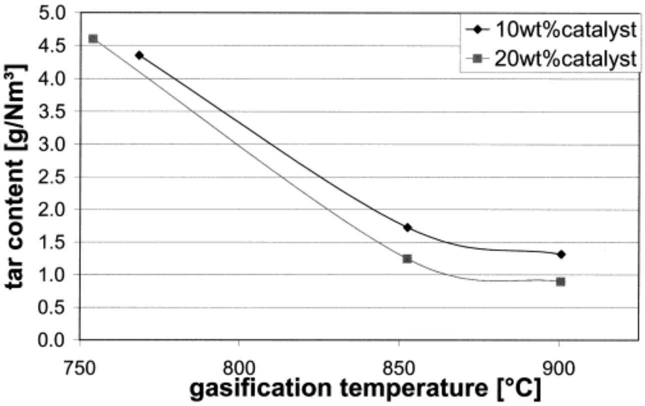

Figure 6. Tar content as a function of gasification temperature and amount of catalyst.

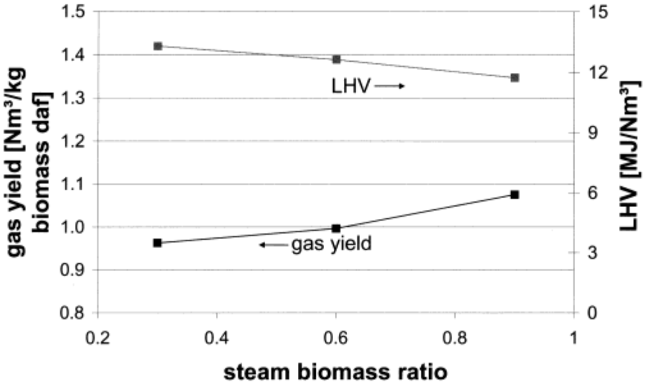

Figure 7. Influence of the steam/fuel ratio on the gas yield and the lower heating value (gasification temperature = 850 °C, 20 wt % catalyst).

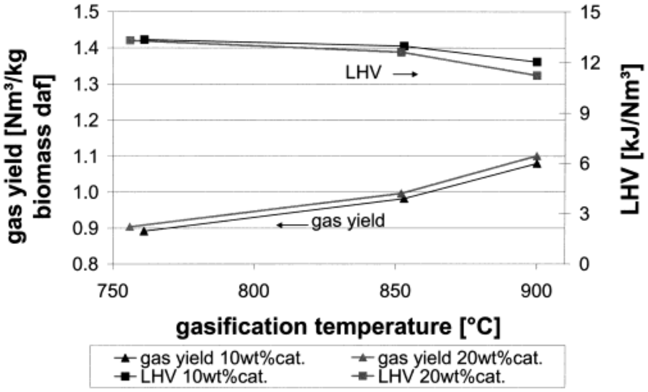

Figure 8. Influence of the gasification temperature and amount of catalyst on the gas yield and the lower heating value (steam/fuel ratio = 0.6).

Figure 6 shows the dependency of the tar content in the dry product gas on the gasification temperature and the amount of catalyst used in the bed material. The tar content is heavily dependent on the temperature. The reason for this effect is that the catalyst shows an increased activity at higher temperatures. From the literature, it is known that the temperature range for catalytic reactions should be from 800 to 900 °C to obtain high conversion rates (see, e.g., ref 16). The marked influence of the temperature explains the higher tar content with 20% catalyst in the bed at 754 °C compared to 10% catalyst in the bed at 767 °C. As already mentioned above, the tendencies concerning temperature are the same with and without the use of a catalyst in the bed; however, the observed dependencies are stronger with the catalyst.

Lower Heating Value. The heating value is the sum of the products of the single volumetric components multiplied by their respective lower heating values.

Higher steam-to-biomass ratios lead to decreased lower heating values (Figures 7 and 8). Increasing the amount of catalyst in the bed material decreases the

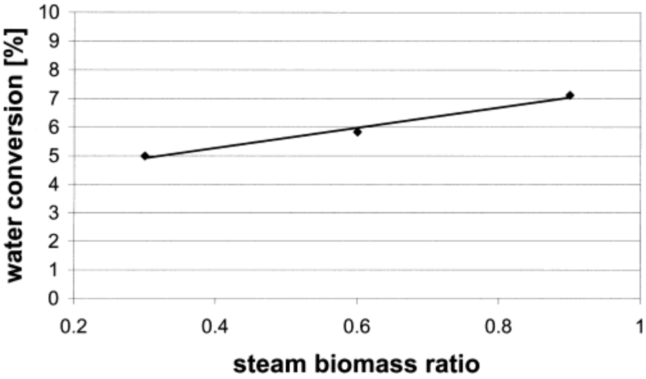

Figure 9. Influence of the steam/fuel ratio on the water conversion (gasification temperature = 850 °C, 20 wt % catalyst).

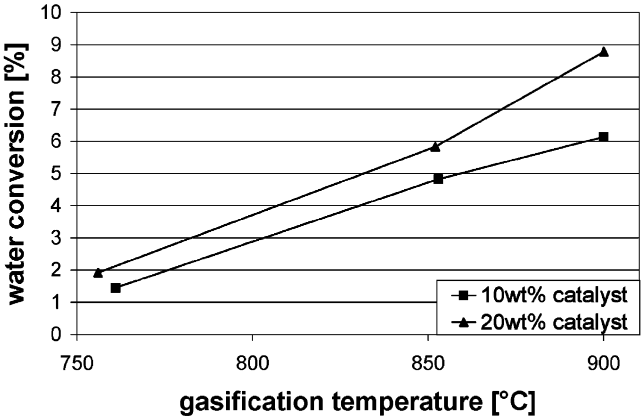

Figure 10. Influence of the gasification temperature and the amount of catalyst on the water conversion (steam/fuel ratio = 0.6).

lower heating value. These trends are the same as at higher temperature (Figure 8). Generally, the lower heating value is decreased by the use of the catalyst because of the higher content of hydrogen (10 783 kJ/Nm^3) and the lower content of methane (35 883 kJ/Nm^3) in the product gas. On the other hand, this effect is compensated by an increased gas yield (Figure 8).

Mass and Energy Balances. To calculate the mass and energy balances, simulation software (IPSEpro, version 3.1) was used. IPSEpro is an equation-oriented stationary simulation program developed for power plant simulations. Therefore, the basic components of conventional power stations are provided by the standard library of IPSEpro. The thermodynamic properties of water and steam, as well as those of the gases typically involved in combustion (Ar, CO, CO_2, N_2, O_2, H_2O, CH_4, C_2H_6, SO_2, C_3H_8, H_2, H_2S), are also provided by the software. IPSEpro allows one to edit the source code of the standard models, as well as to create new models for special process steps. The following results were obtained from the mass and energy balances.

Water Conversion. Water conversion is an important value used to estimate the efficiency of a steam gasification system. The water conversion was calculated according to the equation

$$C_W = \frac{\dot{m}_{w\_in} - \dot{m}_{w\_out}}{\dot{m}_{fuel\_dry\_in}} \qquad (2)$$

Figures 9 and 10 contain the results of the investigations concerning the water conversion. As can be seen, the water conversion increases with increasing steam-to-biomass ratio (Figure 9). This is mainly due to the higher water input in the case of higher steam-to-biomass ratios. The figures show that the main part of the steam leaves the gasifier unreacted. For this reason, an optimum has to be found between the required gas quality and the heat loss due to unreacted steam. In Figure 10, the dependence of the water conversion on the amount of catalyst in bed and the gasification temperature is shown. As expected, the water conversion increases with increasing temperatures as well as with increasing amounts of catalyst.

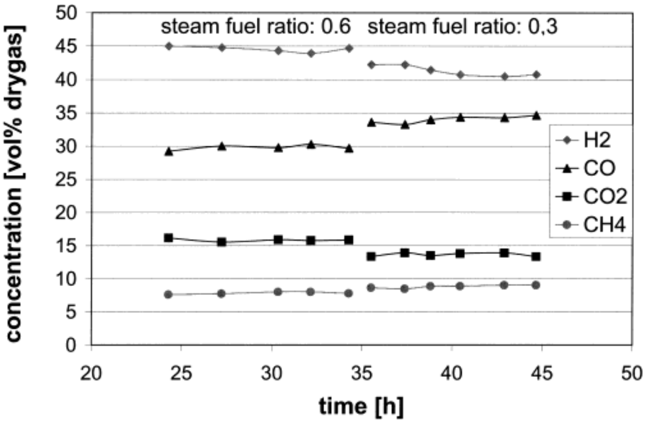

Figure 11. Product gas composition over time (gasification temperature = 850 °C, steam-to-fuel ratio = 0.6 and 0.3, 20 wt % catalyst).

Chemical Efficiency. The chemical efficiency of the gasifier was calculated by dividing the chemical energy of the product gas by the chemical energy of the biomass fuel at the feeding point. In the case of the pilot plant, wood pellets in the gasification zone and additional fuel (light fuel oil) in the combustion zone are used. The following equation was used to calculate the chemical efficiency

$$\eta_{chem} = \frac{LHV_{pg}\dot{V}_{pg}}{LHV_{fuel}\dot{m}_{fuel} + LHV_{add\_fuel}\dot{m}_{add\_fuel}} \qquad (3)$$

In all cases, the chemical efficiency lies within the range of 60−70%. No correlation between the amount of catalyst and the chemical efficiency could be found.

Gas Yield. Higher steam-to-biomass ratios lead to increasing water conversion rates and also to increasing gas yields (Figures 8 and 7). As the amount of catalyst in the bed material is increased, the gas yield is increased slightly, whereas the lower heating value is decreased, as discussed above. The gas yields are higher at higher temperatures with the same trends as the lower heating value (Figure 8).

Lifetime of the Catalyst. The main causes of deactivation of transition metal catalysts are coke deposition and deactivation by sulfur.[22] The coke deposit on the bed material can be removed by burning off the coke in the combustion zone. Furthermore, attrition is important for in-bed catalysts in fluidized bed processes. A comparison of the attrition of pure olivine and olivine and catalyst mixtures showed no significant difference. The attrition rates were about 0.015 kg/kg of fuel (dry). The pneumatic transport in the combustion zone is suspected as the main reason for the attrition of the bed material, because of the higher gas and particle velocities. It is well-known from the literature that the attrition of bed material in a fluidized bed is directly proportional to the gas and particle velocities.[23−26]

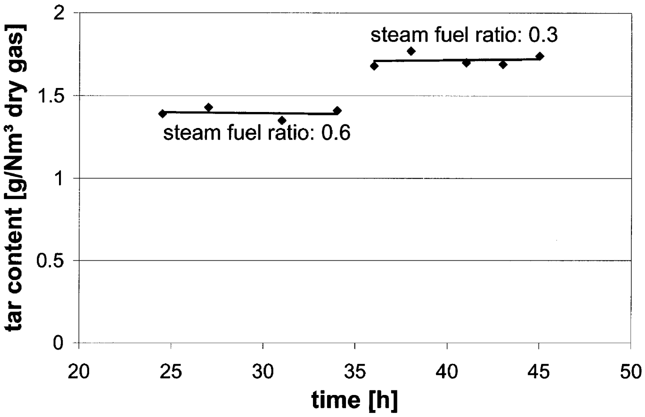

Figure 12. Tar content as a function of time (gasification temperature = 850 °C, steam-to-fuel ratio = 0.6 and 0.3, 20 wt % catalyst).

The catalyst showed no loss of activity over the experimental times of 30 and 45 h. In Figure 11, the dry product gas composition measured using 20 wt % of catalyst in the bed material is shown as a function of time. This experiment lasted for 45 h (gasification time excluding the start-up and shut-down periods, which last 3 h each) and was carried out at reference conditions (gasification temperature of 850 °C, steam-to-fuel ratios of 0.6 and 0.3). After the steam-to-fuel ratio had been changed, it took about 1 h to return to constant process conditions inside the gasifier (temperatures). The gas composition became constant after about 4 h after the steam-to-fuel ratio had been changed. It is evident that the catalyst experienced no loss of activity over the experimental time. This is also indicated by the tar content in the product gas over time, which is shown in Figure 12. Higher steam-to-fuel ratios led to higher tar contents, as was found in previous experiments with olivine alone.[21]

The same result was found during the second long time test of over 30 h using 5 wt % of catalyst in the same experimental setup. These results were similar to those obtained in earlier investigations using 43 wt % of catalyst at a gasification temperature of 800 °C and a steam-to-fuel ratio of 0.58.

Conclusions

Improving gas cleaning technologies is definitely important for the technical feasibility of fluidized bed gasification processes. The investigations described herein that were performed at a pilot plant showed that the catalyst exhibits a promising ability to improve the quality (low tar content and high hydrogen volume fractions) of the product gas of the so-called FICFB (fast internally circulating fluidized bed) gasification process. With different mixtures of catalyst and natural olivine (0−43%), good performances of the catalyst were observed.

Generally, the catalyst exhibits the same fluid dynamic behavior as natural olivine and is therefore well-suited to be used as a bed material in fluidized beds. Thus, the catalyst can be used in the FICFB gasification process in different mixtures with natural olivine without any separation effects during operation of the gasifier and can even be used alone.

The catalyst showed a high activity in the steam reforming of methane and tars. The tar content of the product gas could be reduced by up to 75% while the hydrogen volume fraction could be increased by up to 8 percentage points compared to natural olivine as the bed material.

During the experiments, no measurable influence of the amount of catalyst in the bed material on the chemical efficiency was detected. An increase in the temperature or the amount of catalyst resulted in an increase in the water conversion, as well as in the tar reforming. The gas yield followed the same tendencies. In contrast, the lower heating value of the product gas decreased.

Independently of the ratio between the catalyst and the natural olivine in the bed material, the measured attrition was 0.015 kg/kg of fuel (dry). During the long-duration experiments (up to 45 h), no deactivation of the catalyst could be observed. Of course, this time on stream is not sufficient for a commercial application, and further testing of this catalyst in long-term tests is required. To this point, the problem of the cleaning the product gas from biomass gasification has not been completely solved. Catalytic tar removal seems, as mentioned in the Introduction and supported by the results in this paper, the best way to clean the product gas from polyaromatic hydrocarbons. The nickel-based catalyst used here destroys tars present in the gas in a very efficient way. Putting the catalyst in the fluidized bed is a good solution that enhances the efficiency of the process, simplifies the gas cleaning line, and decreases the operating costs.

Acknowledgment

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Nomenclature

C_{w} = water conversion
HHV = higher heating value of the wood pellets (kJ/kg)
$\mathrm{LHV}_{\mathrm{add\_fuel}}$ = lower heating value of the additional fuel (kJ/kg)
$\mathrm{LHV}_{\mathrm{fuel}}$ = lower heating value of the wood pellets (kJ/kg)
$\mathrm{LHV}_{\mathrm{pg}}$ = lower heating value of the product gas (kJ/Nm^3)
$\dot{m}_{\mathrm{add\_fuel}}$ = mass flow of the additional fuel (kg/h)
$\dot{m}_{\mathrm{fuel}}$ = mass flow of the wood pellets (kg/h)
$\dot{m}_{\mathrm{fuel\_dry\_in}}$ = mass flow of the dry biomass (kg/h)
$\dot{m}_{\mathrm{w\_in}}$ = total mass flow of the water in the gasifier (kg/h)
$\dot{m}_{\mathrm{w\_fuel\_in}}$ = mass flow of the water in the gasifier with the biomass (kg/h)
$\dot{m}_{\mathrm{w\_fluid\_in}}$ = mass flow of the water for the fluidization (kg/h)
$\dot{m}_{\mathrm{w\_out}}$ = total mass flow of the water in the product gas (kg/h)
sfr = steam-to-fuel ratio
T_{B} = boiling temperature (°C)
V_{pg} = volume flow of the product gas (Nm^3/h)
η_{chem} = chemical efficiency of the gasifier (%)

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