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ORIGINAL ARTICLE

Internal tar/CH_4 reforming in a biomass dual fluidised bed gasifier

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Abstract An internal reformer is developed for in situ catalytic reforming of tar and methane (CH_4) in allothermal gasifiers. The study has been performed in the 150 kW dual fluidised bed (DFB) biomass gasifier at Mid Sweden University (MIUN). The MIUN gasifier is built for research on synthetic fuel production. Reduction of tars and CH_4 (except for methanation application) in the syngas is a major challenge for commercialization of biomass fluidised-bed gasification technology towards automotive fuel production. The MIUN gasifier has a unique design with an internal reformer, where intensive contact of gas and catalytic solids improves the reforming reactions. This paper presents an initial study on the internal reformer operated with and without Ni-catalytic pellets, by evaluation of the syngas composition and tar/CH_4 content. A novel application of Ni-catalyst in DFB gasifiers is proposed and studied in this work. It can be concluded that the reformer with Ni-catalytic pellets clearly gives a higher H_2 content together with lower CH_4 and tar contents in the syngas than the reformer without Ni-catalytic pellets. The gravimetric tar content decreases down to 5 g/m^3 and the CH_4 content down below 6 % in the syngas. The tar content can be decreased further to lower levels, with increased gas contact to the specific surface area of the catalyst and increased catalyst surface-to-volume ratio. The new design in the MIUN gasifier increases the gasification efficiency, suppresses the tar generation and upgrades the syngas quality.

Keywords Biomass gasification · Syngas cleaning · Tar removal · Tar/CH_4 reforming · Dual fluidised bed · Ni-catalyst

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1 Introduction

The 1st generation of biofuels for transportation is no longer encouraged as it is usually difficult to meet criteria of greenhouse gas (GHG) saving when replacing fossil fuels. On the other hand, the 2nd generation of biofuels from lignocellulosic biomass has been more and more attractive for development and commercialization. Transport biofuels as well as chemicals can be produced from high-quality syngas (mainly hydrogen and carbon monoxide) via biomass gasification [1].

When syngas is used for synthesis, a high CO + H_2 concentration (>80 %) with a high H_2/CO ratio in the syngas is required to ensure smooth downstream synthesis. In addition to H_2 + CO, the raw syngas from biomass gasifiers contains CH_4, trace amounts of higher hydrocarbons (tars), CO_2, possible inert gases from biomass and gasification agent and various contaminants. There are many years of experience in gas cleaning related to engine and turbine applications, but product gas for synthesis normally has a much stricter specification of impurities than these applications [2]. Syngas can be conditioned to different degree depending on a balance of economic cost against technical specification for downstream synthesis. The downstream syngas cleaning usually accounts for up to 38 % of the transport fuel production cost [3]. The major challenge in the production of high quality syngas through biomass fluidised-bed gasification is the reforming of tars and methane (except for methanation application) to a minimum allowable limit.

Reduction of tars and methane (CH_4) to an acceptable low level is usually achieved by high temperature thermal cracking, low temperature catalytic cracking, or physical tar treatment like water scrubbing + sedimentation and oil scrubbing + combustion [2, 4–8]. Catalytic cracking efficiency can be 90–95 % at reaction temperatures about 800~900 °C [9], whereas thermal cracking requires temperatures above 1200 °C to reach the same efficiency at

expense of energy losses and big investments on high temperature materials.

The catalysts can be used as the secondary method, in downstream catalytic reactors [10, 11], such as catalytic beds, monoliths and filters, or as the primary method, e.g. added directly in the fluidised bed gasifier as the bed material. The use of catalytically active bed materials promotes char gasification, water-gas-shift (WGS) and steam reforming reactions, which can enhance tar/CH_4 reforming and increase the H_2 content in the syngas. The primary method is more cost-effective attributed to lower thermal losses, less downstream reactors and lower investment cost.

Ni-supported olivine is highly effective in reduction of tars and CH_4 [12], but an important drawback is the toxicity of nickel and the volatile particles that occurs in fluidised bed gasifiers. Commercial Ni-catalysts have been widely tested as secondary catalysts for tar reduction and methane reforming [13–17]. The application of Ni-catalyst in DFB gasifiers is emphasized in this work. The catalyst is usually deactivated in a gasification reactor due to carbon deposition, chloride, sulphur poisoning, oxidation and sintering. However, the lifetime of the catalyst can be prolonged by the oxygen balance in a dual fluidised bed gasifier (DFBG) [11, 12].

One common catalytic bed-material used in DFBGs is olivine ((Mg, Fe)2 SiO4), a natural mineral containing magnesium, iron oxide and silica. Catalytic activity of olivine in cracking and reforming of tars and enhanced steam and dry reforming of hydrocarbons are reported in a number of articles [18, 19]. Olivine has been a more and more attractive bed material used in DFBGs due to good attrition resistance comparable to silica sand, low price and good catalytic effect comparable to dolomite. However, the tar/CH_4 reforming performance by the olivine or Fe-doped olivine catalytic bed materials is still limited [20]. The performance might be improved by more intensive contact between volatile gas and catalytic bed material at a higher temperature zone in the gasifier, above the dense bed where the hot bed material returning from the combustor.

Different approaches similar to above mentioned ideas have been suggested by a few researchers. One example is the two-stage DFBG, proposed by Xu and his colleagues in 2008 [21]. This design uses the principle of two fluidised beds without external upper pressure lock. Another example is the G-volution design concept proposed by Schmid et al. [22] with integrated flow obstacles to create a counter current flow pattern to increase gas-solid contact.

A 150 kW DFBG was built at Mid Sweden University (MIUN) in 2007 [23] which has a unique design suitable for in-bed tar/CH_4 catalytic reforming and continuously internal regeneration of the reactive bed material. The catalytic effects of calcined olivine and Fe-doped olivine (10 % wt Fe-olivine catalyst synthesized by impregnation of an aqueous iron nitrate solution.) have been studied in the MIUN gasifier [20].

Fig. 1 The 150 kWth MIUN biomass DFBG gasifier

The results show fairly high methane and gravimetric tar contents in the raw syngas (~10 vol.% and 7–67 g/m^3, respectively). In order to further reduce the tar and CH_4 contents in the syngas, an in situ reformer referred to as *FreeRef* reformer [24] is developed and installed in the freeboard of the fluidised bed gasifier. This work presents an experimental study on FreeRef reformer for in situ catalytic reforming of tar and CH_4.

Table 1 Fuel analysis of wood pellets (6 mm) produced by SCA BioNorr AB, Sweden

Elementary analysis	% db	
Oxygen O (calc.)	42.7	
Sulphur S	<0.01	
Chlorine Cl	<0.01	
Carbon C	50.9	
Hydrogen H	5.9	
Nitrogen N	<0.1	
Ash cont.	0.4	% db
Moisture	6.5	%
Durability of pellets	98.1	%
Bulk density	667	kg/m^3
Net calorific value as rec.	18.849	MJ/kg
Net calorific value db	20.159	MJ/kg

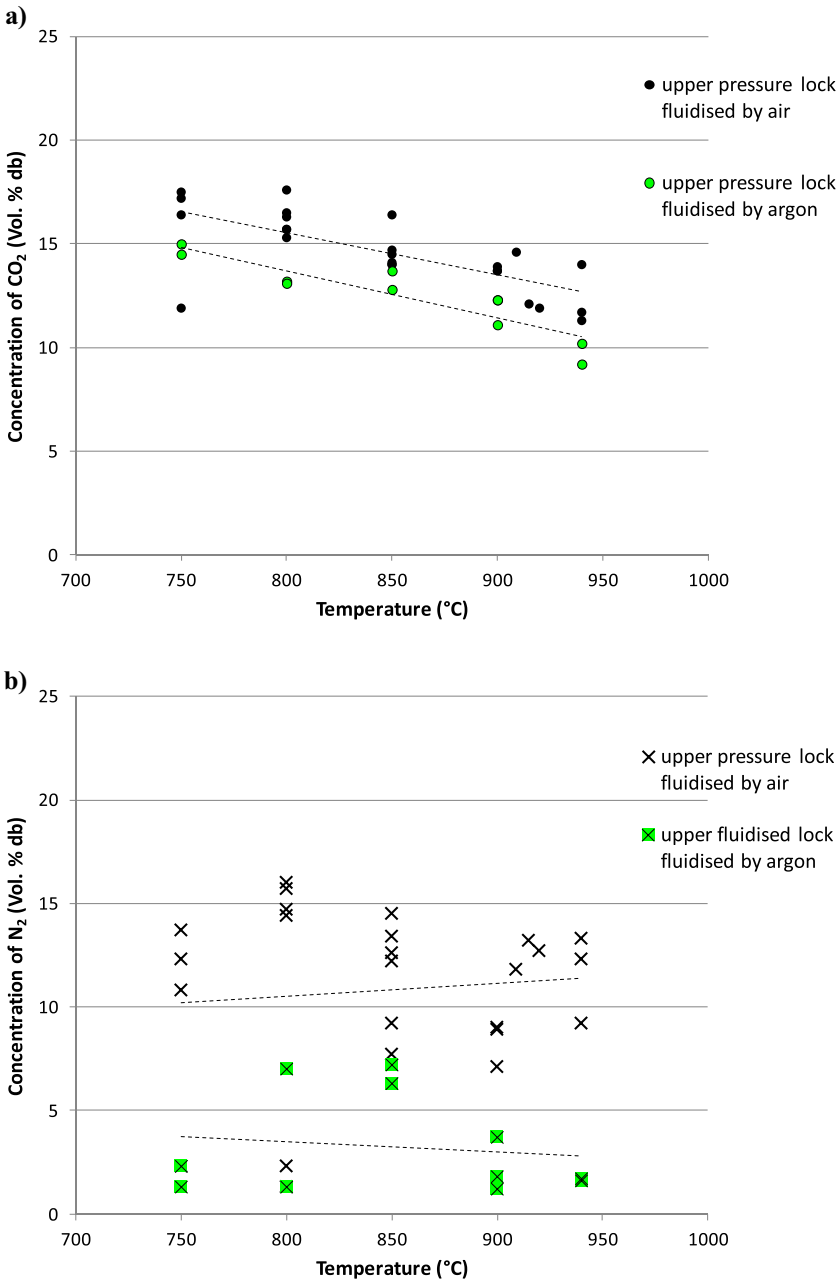

Fig. 2 The concentrations of (**a**) CO_2 and (**b**) N_2 in the syngas when air or argon is used to fluidise the upper pressure lock

2 Experimental setup

2.1 MIUN gasifier

The MIUN gasifier is a DFBG (see Fig. 1) and consists of a bubbling fluidised bed (BFB) steam gasifier and a circulating fluidised bed (CFB) riser combustor, and has the biomass treatment capacity of 150 kWth, i.e. approx. 25 kg biomass feed per hour. The feedstock is wood pellets (see Table 1). The gasifier and the combustor have a height of 2.5 and 3.1 m and inner diameters (i.d.) of 300 and 90 mm, respectively. The MIUN gasifier has been described in detail in a previous article [23].

The bed material in these tests is olivine. The oxygen transport capacity of olivine can be 0.5 wt.% [25]. Hence, the produced gas in the gasifier will be partially oxidized by oxygen input by the olivine in dual fluidised beds (DFB) operation. Reduction of bed material in the steam gasifier with a following oxidation in the air combustor achieves a catalyst regeneration cycle, similar to the chemical looping combustion (CLC) [26]. The olivine is pre-treated by calcination to increase the free iron (III) concentration on the olivine surface for better catalytic activity. The olivine in these tests is calcined inside the DFB reactor at 900 °C in 10 hours, with air at slightly elevated pressure [27].

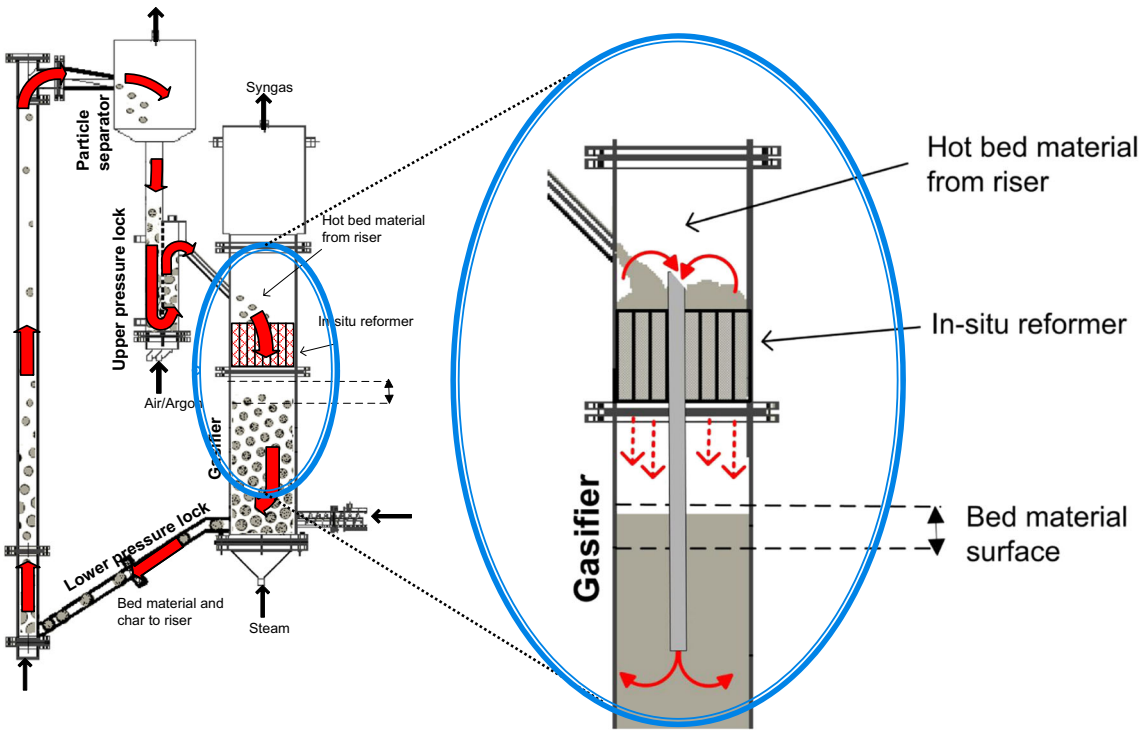

Fig. 3 MIUN DFBG gasifier with in situ reformer

In the experiment, the gasifier is fluidised with steam and the riser with air. Due to operation convenience, the upper pressure lock is fluidised with air instead of steam. Likewise, air is used for aeration of the lower pressure lock if necessary.

Reference tests are performed, in which air to the upper pressure lock is changed to Argon, to investigate how the fluidisation air to the upper pressure lock affects the syngas. Figure 2 shows the contents of N_2 and CO_2 in the syngas as a function of gasification temperature, when the pressure lock is fluidised with air or argon, respectively. When air is used, the content of N_2 is around 12 % in volume and CO_2 15 %. With Argon, the content of N_2 is about 3 % and CO_2 13 %, which can be attributed to air introduction from the feedstock, the feeding system and possible air leakage from the combustor into the gasifier. When the upper pressure lock is fluidised with air, the CO_2 content in the syngas is increased (see Fig. 2a), due to oxidation reactions. This suggests a minor influence of the air introduction from the upper pressure lock on the syngas composition.

2.2 In situ reformer

In general, the biomass gasification process occurs through three steps: (1) pyrolysis which devolatilises biomass into char and volatile matter including tars; (2) secondary reactions such as cracking and reforming of tars and (3) gasification reactions of the remaining carbonaceous residue with steam and carbon dioxide. The steps 1 and 3 take place in the dense bed of the fluidised bed gasifier, while step 2 mainly takes place in the freeboard of the fluidised bed. Thus, the reforming of tars and CH_4 in the freeboard is important for high quality syngas production, which is favoured by a good contact of volatile with catalytic bed material at a sufficiently high temperature. This might not be the case for the freeboard region of MIUN gasifier, where the hot bed material returns next to the wall while the volatile passes through the centre. This insufficient gas-solid contact may be one reason that the tar content in the syngas in previous test of MIUN gasifier [20] is not low enough, although the bed material Fe-olivine was used as the circulating catalytic bed material.

A good gas-solid contact can be realized by an in situ reformer installed above the dense bed of the fluidised bed gasifier but under the hot bed material return position. Such an in situ reformer, similar to an upper fluidised bed of olivine, is developed in this work for MIUN gasifier as shown in Fig. 3. The *FreeRef* reformer is a kind of an upper stage fluidised bed.

Table 2 Physical and chemical properties of the steam reforming catalytic pellets

Appearance	5-hole drums
Particle size (mm)	19–20
Pore diameter (mm)	3–5
Bulk density (kg/l)	0.90–0.95
Crushing strength (N/particle)	>300
NiO (%)	>14
Free silica (%)	0.2

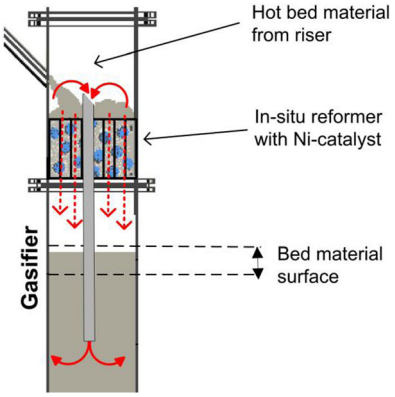

Fig. 4 Ni-based catalytic pellets for steam reforming of tars and CH_4

The hot bed material passes through the reformer and creates a fluidisation region, with the highest temperature in the gasifier. The main part of the bed material overflow via the centrum pipe, and a minor flow of the bed material flows through the distributor of the fluidised bed reformer, on the way down to the bottom bed of the gasifier to provide heat for biomass pyrolysis. This flow pattern has been studied in an acrylic cold flow model to determine the design. The volatile from biomass pyrolysis, on the other hand, flows through the in situ reformer and experiences cracking and reforming of tars and CH_4 in the hotter reformer.

2.3 In situ reformer with Ni-catalytic pellets

In this test, the reformer is filled with Ni-based catalytic pellets (see Table 2 and Fig. 4). Nickel oxide is the active component with alumina as support material. The catalyst has good stability and high mechanical strength, and can be used at 1500 °C for a long time.

The in situ reformer enables regeneration/cleaning of the catalytic surface from contaminants due to some abrasion from the circulated bed material keeping the pores open. In addition, when utilizing an oxygen carrier as bed material, the oxides introduced by the bed material to the in situ reformer promotes regeneration of the catalytic properties due to oxidizing of substances such as sulphur and carbon.

2.4 Analysis of gas composition and tars

The main syngas stream from the gasifier is led to an incinerator for complete combustion. A slip stream of the syngas passes through the gas sampling and analysis system. Electrical heaters are used to keep the syngas temperature at about 400 °C to avoid tar condensation in the pipes. For the measurements of gas composition, the syngas is subtracted by a vacuum pump and sampled manually in Tedlar gas sampling bags to be analyzed off-line in a parallel FID and TCD GC-detection system. The gas was sampled four times in each experimental test under the same gasifier operation condition. Gas contents H_2, CO, CH_4, CO_2 and O_2 are also analyzed on-line with an *X-STREAM Enhanced XEGP—General Purpose Gas Analyzer*.

Most heavy tars are GC-undetectable, and here, they are gravimetrically analyzed. The results include GC-undetectable heavy tar compounds together with some GC-detectable tars (from 2 to 3 rings) [28] in addition to all compounds larger than 3 rings [29]. The gas passes a high temperature filter (*Munktel ET/MK 360* Quartz thimble 34×150 mm, penetration <0.002 % DOP (0.3 μm)) held at 400 °C during sampling and cooled down to 25 °C while passing through two tar capture glass fibre filters in series (*Whatman GF/D*, particle retention 2.7 μm, thickness 675 μm at filtering speed of about 16.5 ml/s (see Fig. 5). It took about 45 s. to collect 4-l product gas from the gasifier. After each sampling, the glass fibre-filter adapters in the tar collector are washed with isopropanol. Finally, the tars are collected in a round flask containing isopropanol. The detailed sampling procedure can be found elsewhere [23].

The solution containing the tar sample together with some possible filter fibres is filtered through a glass fibre funnel (pore size 10–16 μm) and collected to a new round flask that has been weighted ahead.

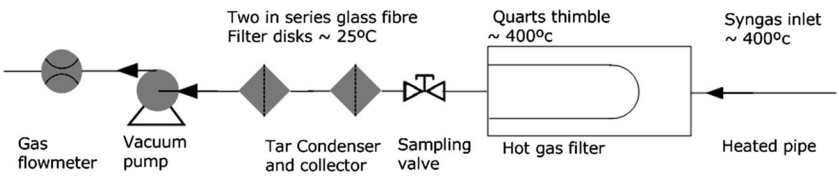

Fig. 5 Gravimetric tars sampling system (the syngas passes through two tar capture glass fibre filters in series at ambient temperature whereas the tars condense)

Finally, the round flask is inserted into an evaporation condenser. When the tar sticks on the inside of the round flask, the evaporation is finished. The weight difference, i.e. the tar content, can finally be calculated. The analytical balance applied is a *Precisa XR 205SM-DR*, readability 0.01 mg/0.1 mg, with a built-in self-calibration system. The remaining tars are referred to as "gravimetric tar".

2.5 Experimental test

In this study, 13 gasification tests were performed at the gasifier operation temperature from 750 to 940 °C in the two cases with and without Ni-catalytic pellets in the in situ reformer (see Table 3). The circulating bed material is calcined olivine with Sauter mean particle size of 223 μm. The biomass feedstock is wood pellets. The steam-to-carbon ratio is 1.2 and is calculated according to Eq. (1).

$$S/C = \frac{\dot{m}_{\text{steam}} + \nu_{H_2O} \times \dot{m}_{\text{biomass}}}{\nu_C \times \dot{m}_{\text{biomass}}}$$

where $\dot{m}_{\text{steam}}$ represents the mass flow of steam (kg/s)

$\dot{m}_{\text{biomass}}$ represents the flow of biomass (kg/s)

ν_C represents the carbon mass fraction in the biomass

ν_{H_2O} represents the water mass fraction in the biomass (1)

Each test started after stabilization of the gasification temperature. The gas and tar sampling were carried out when the gasifier had reached steady state condition and at stable syngas composition according to the on-line gas analyzer. The same batch of bed material was used during the whole test series.

The performance of the in situ reformer is evaluated based on the temperature distribution in the gasifier, the syngas composition, H_2/CO ratio, CH_4 concentration and the gravimetric tar content in the syngas. The two cases of DFB with an in situ reformer with and without filling of Ni-catalytic pellets are compared to BFB and DFB that were studied previously [20] regarding the syngas quality. Electrical heaters allow separate operation of the steam gasifier as a BFB gasifier. At BFB operation, the interconnections between the gasifier and the riser are blocked and hence are the vessels divided. The only heat source for BFB operation is the electrical heaters.

3 Results and discussion

All measurement data of syngas composition and tar content are summarized in Table 3 in average value for each gasification test on dry basis. The off-line measured values correspond well with the registered values from the on-line gas analyzer. The reference test shown in Fig. 2 indicates that the impact of air from the upper pressure lock on the gas composition and the gravimetric tar content is limited, which can also been seen in Table 3.

The in situ reformer filled with Ni-catalytic pellets results in a clear increase in the H_2 concentration of the syngas, together with clear reduction of gravimetric tar and methane contents.

Table 3 Measurement results (mean values) of the test in the MIUN gasifier (S/C=1.2)

Test:	1	2	3	4	5	6	7	8	9	10	11	12	13
Temp. (°C):	750	800	850	900	940	750	800	850	900	909	915	920	940
In-Situ Reformer:	Without Ni-catalytic pellets					With Ni-catalytic pellets							
Gas comp.(Vol.% db):													
H_2	29.8	28.7	31.8	35.4	34.4	24.8	28.8	34.2	38.1	35.5	40.4	40.7	41.5
CO	31.7	31.9	34.9	34.7	35.8	28.2	28.1	28.3	32.8	30.6	33.5	33.1	32.4
CH_4	11.0	10.6	11.4	10.1	9.0	10.7	9.4	7.5	6.8	6.7	5.9	6.2	5.7
CO_2	15.5	15.0	14.0	13.2	11.0	16.8	15.6	14.2	11.1	12.1	11.9	12.0	10.2
Ethene	4.1	4.0	4.1	3.2	2.5	3.4	3.1	2.4	1.5	1.8	1.6	1.2	0.6
Ethane	0.6	0.4	0.3	0.1	0.0	0.6	0.5	0.3	0.1	0.2	0.3	0.1	0.1
C3	0.2	0.1	0.1	0.0	0.0	0.3	0.2	0.1	0.0	0.0	0.0	0.0	0.0
C4	0.1	0.0	0.2	0.0	0.1	0.1	0.1	0.1	0.0	0.0	0.0	0.0	0.0
O_2	0.8	0.7	0.4	0.9	0.9	0.7	0.8	1.5	N/A*	0.9	0.9	0.7	N/A*
N_2*	12.3	15.0	8,0	9.0	12.1	14.7	12.7	12.6	3.7*	6.8	6.2	5.9	1.7*
H_2+CO (Vol.% db):	61.5	60.6	66.7	70.1	70.2	53.0	56.9	62.5	70.9	66.1	73.9	73.8	73.9
H_2/CO	0.9	0.9	0.9	1.0	1.0	0.9	1.0	1.2	1.2	1.2	1.2	1.2	1.3
Grav. Tar (g/Nm^3):	23.2	13.3	26.0	12.5	16.3	23.7	14.5	10.6	6.3	6.3	5.9	5.0	5.1.

[a] Upper pressure lock fluidised with Argon instead of air

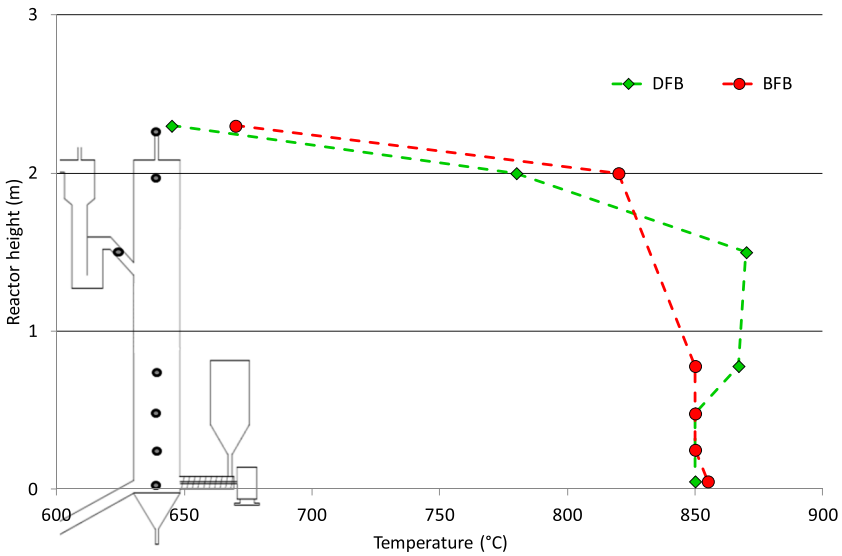

Fig. 6 Temperature profiles of the MIUN gasifier over reactor height in BFB and DFB modes

3.1 Temperature profile over the height of the gasifier

The temperature profiles of the MIUN gasifier over the reactor height in previous tests in BFB and DFB modes are shown in Fig. 6. In the DFB mode, the recycling of hot bed material gives rise to a hot spot in contrast to the BFB mode. The temperature profiles for the DFB after installation of the in situ reformer with and without Ni-catalytic pellets are depicted in Fig. 7. The temperature in the dense bed of the gasifier is fairly constant in spite of biomass feeding above and steam injection under the distributor.

The hottest region up to 900 °C of the in situ reformer can be seen for the case without Ni-catalytic pellets, which can be well-explained by the heating-up from the hot circulating bed material. The in situ reformer has a uniform temperature due to the characteristics of intensive heat and mass transfers in fluidised beds. But for the in situ reformer with Ni-catalytic pellets, the temperature drops down at the bottom level of the reformer. This indicates a strong endothermic reaction of steam reforming of tars and CH_4 by use of Ni-catalyst. The cold spot close to the bottom of the reformer can be attributed to an interaction of two effects, the cooling by the endothermic reforming of tars and CH_4 and the heating by the hot circulating bed material.

3.2 Main gas components versus temperature

Figures 8, 9, 10, and 11 show main gas components as a function of the gasification temperature for comparison of two cases, with or without Ni-catalytic pellets filled in the in situ reformer. It can be seen from the figures that higher

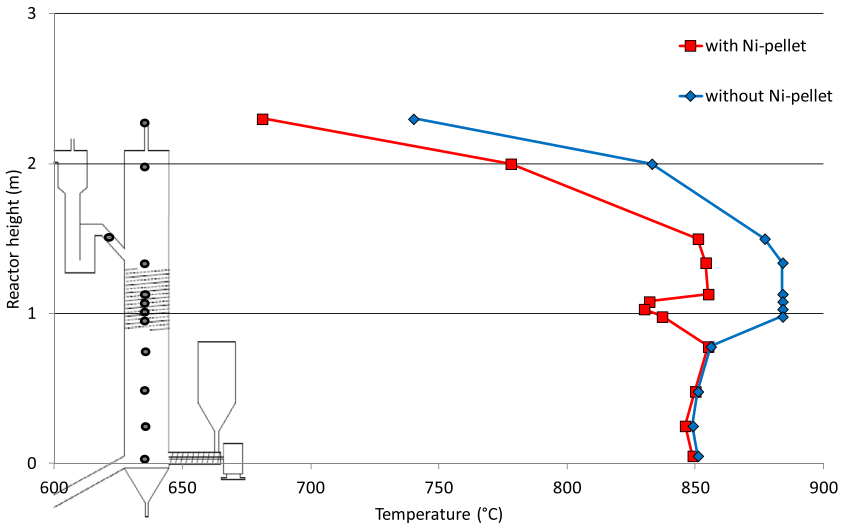

Fig. 7 Temperature profile of the MIUN gasifier with an in situ reformer for two cases, with and without Ni-catalytic pellets

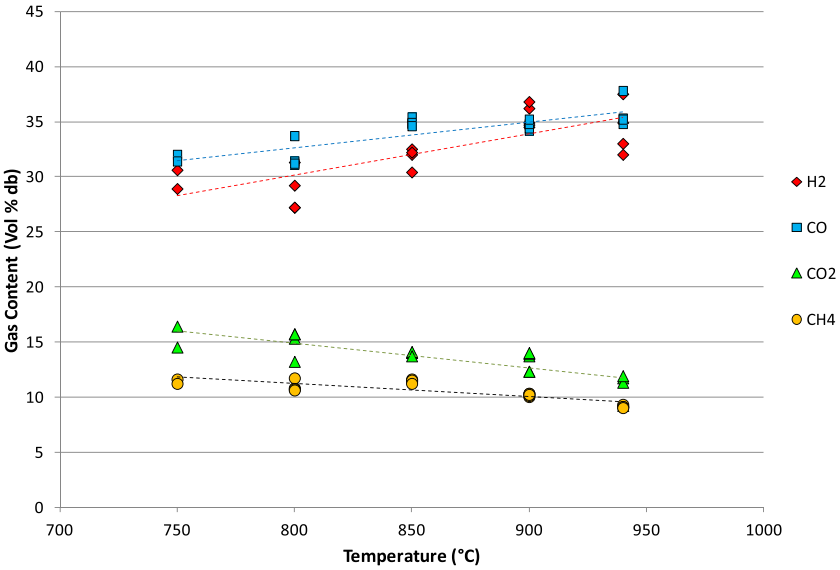

Fig. 8 Main gas components of syngas from MIUN gasifier with in situ reformer

temperature enhances the tar/CH_4 steam reforming reactions and results in higher content of H_2 and CO, while the CO_2 content slightly decreases since the exothermic shift reaction is favoured by low temperature. The contents in CO_2 and CH_4 are decreasing with temperature.

As seen in Table 3 and Figs. 8 and 9, the CO + H_2 concentration clearly increases with temperature from 60 to 70 % for the case without Ni-pellets and from 55 to 74 % for the case with Ni-pellets. The most significant change is H_2 concentration when Ni-pellets are added in the in situ reformer, which increases from 24.8 % at 750 °C to 41.5 % at 940 °C and exceeds the CO concentration at 780 °C. The Ni-pellets hardly change the CO concentration as shown in Table 3. These lead to a H_2/CO ratio under 1 for the reformer without Ni-pellets and above 1 for the reformer with Ni-pellets as seen in Table 3 and Fig. 10.

The H_2/CO ratio varies from 0.9 to 1.0 for the reformer without Ni-pellets and from 0.9 to 1.3 for the reformer with Ni-pellets. A clear trend of H_2 concentration increasing with temperature, suggests a strong steam reforming of hydrocarbons in the in situ reformer by the Ni-catalyst.

Steam reforming of CH_4 is a strongly endothermic reaction:

$$CH_4 + H_2O \leftrightarrow CO + 3H_2 \quad \Delta H°298 = +206\ \mathrm{kJ/mol} \tag{2}$$

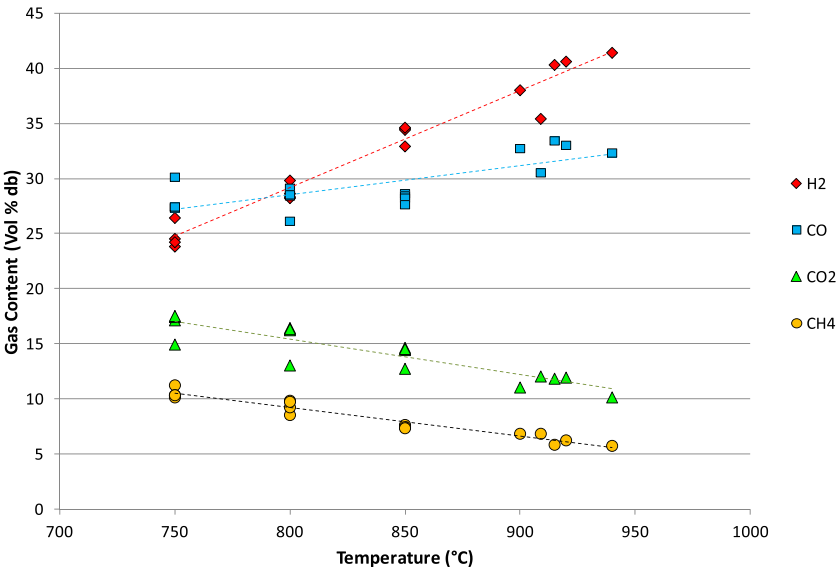

Fig. 9 Main gas components of syngas from MIUN gasifier with in situ reformer when Ni-catalytic pellets is added

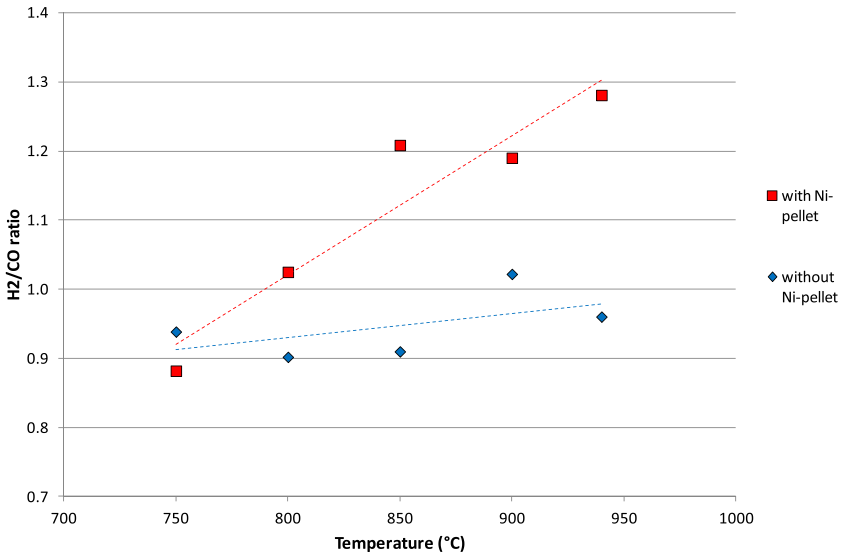

Fig. 10 The H_2/CO ratio (mean values) of syngas from MIUN gasifier with in situ reformer

Under the condition of the in situ reformer, the moderately exothermic water-gas shift (WGS) reaction is extremely fast leading to the equilibrium state:

$$CO + H_2O \leftrightarrow CO_2 + H_2 \quad \Delta H°298 = -41\ \mathrm{kJ/mol} \quad (3)$$

Steam reforming of hydrocarbons is favoured by high temperature; in contrast, the exothermic shift reaction is favoured by low temperature. The amount of steam will enhance the CH_4 conversion. The syngas composition is thus governed by the reactions (2) and (3) above, including reforming of other hydrocarbons. Figure 11 shows the CH_4 concentration. CH_4 is the most recalcitrant hydrocarbon to reform, which very much depends on the temperature as also shown in Fig. 11. For the in situ reformer without Ni-pellets, a decrease in CH_4 content from 11 to 9 % is found at the higher temperatures. For the in situ reformer with Ni-pellets, the CH_4 content is decreasing more clearly from 11 to 6 %. These results indicate that the in situ reformer with catalytic pellets is active in the steam reforming of hydrocarbons.

3.3 Gravimetric tar content versus temperature

Figure 12 shows that the tar content decreases with temperature as a general trend similar to CH_4. The reformer filled with Ni-catalytic pellets shows a higher reduction of tars than the unfilled reformer. By means of the in situ reformer with Ni-pellets, the gravimetric tars significantly decrease from about 25 g/m^3 at 750 °C down to 5 g/m^3 at approx. 920 °C.

It should be noted, this is the initial test, and hence, neither the properties of the catalytic pellets nor the properties of the reformer are optimal. The catalytic pellets have a comparatively large particle size in relation to the size of the reformer

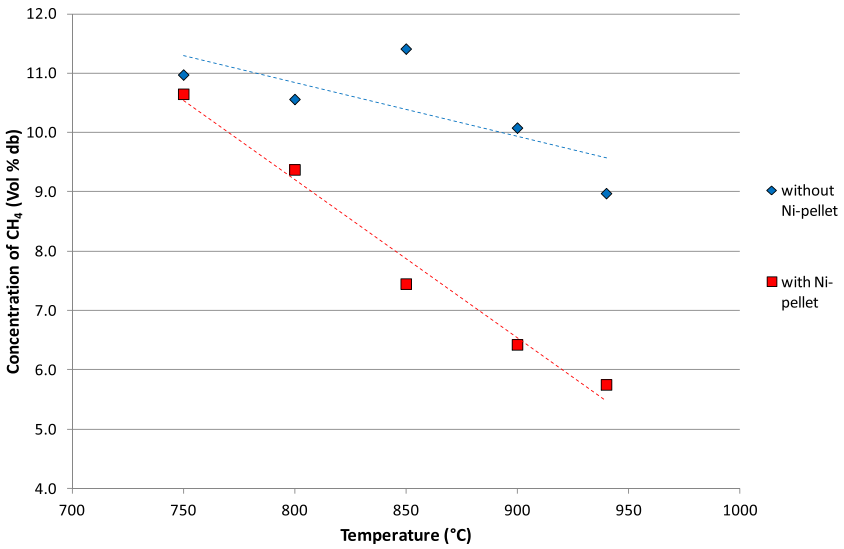

Fig. 11 Methane concentration (mean values) in the syngas from MIUN gasifier with in situ reformer

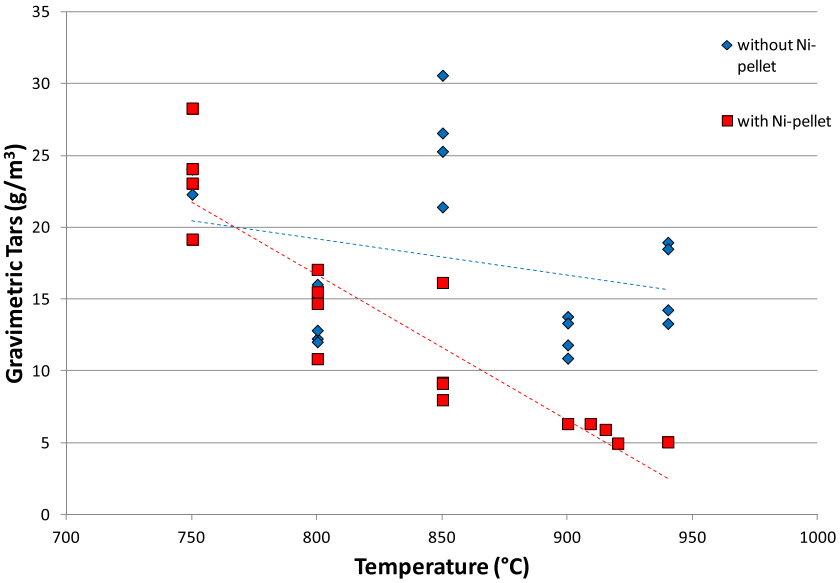

Fig. 12 Gravimetric tar content in the syngas from MIUN gasifier with in situ reformer

vessel. Aznar et.al. evaluated different commercial Ni-based catalyst that reached tar conversions of approx. 98 %. The catalyst was crushed and sieved to a size of - 1.0+0.2 mm [14], which has much smaller particle size than the pellet used in this work.

Thus, the relatively high tar yield of the tests with in situ reformer filled with Ni-catalytic pellets, in comparison with other reported gasification tests with downstream Ni-catalyst reactor (tar yield <2 g/m^3 [30, 31]), can be explained by a low specific surface area of the pellet catalyst, a short gas residence time and an insufficient gas-solid contact The design of the in situ reformer should be optimized to further reduce the tar content in the syngas to 1–2 g/m^3. Then, it's possible to use simplified cleaning equipment such as oil- and water-scrubbers to reach a tar level below 10 mg/m^3 in the syngas.

Commercial Ni-catalysts have been widely tested as secondary catalysts for tar removal and methane reforming. The application of Ni-catalyst in the FreeRef reformer of DFB gasifiers is emphasized in this work. The novel reactor design enables application of catalytic materials directly in the gasification vessel, in spite of high tar content and other contaminants in the inlet gas, since the circulated bed material provides regeneration/cleaning of the active sites on the catalytic surface due to abrasion and oxidation. So far, the measured tar content in the syngas is fairly independent of the experimental time in this study, which indicates little or no deactivation of Ni-catalyst. A detail study on the performance of Ni-catalytic pellets in the in situ reformer of a fluidised bed environment needs to be done further.

3.4 Comparison of DFB gasifiers in different modes

With regard to the main gas components and gravimetric tar content in the syngas, a comparison of four modes of gasifiers

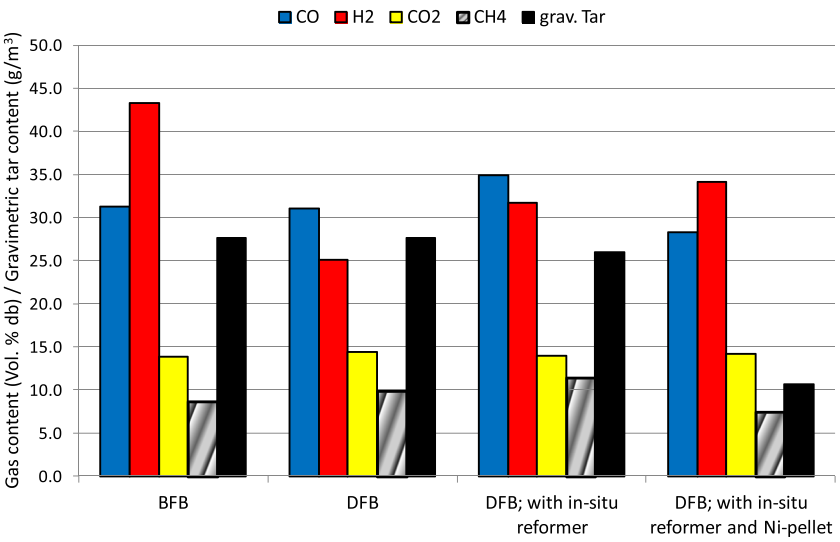

Fig. 13 Mean values of the main gas components and gravimetric tars in the syngas of MIUN gasifier operated at 850 °C

is made: BFB, DFB without in situ reformer, DFB with in situ reformer and DFB with in situ reformer filled with Ni-pellets, and shown in Fig. 13 in averaged values of all tests. The measurement data for BFB and DFB without in situ reformer are taken from previous tests [20].

The concentrations of CO_2 and CO stay at the same level for the four modes, but the H_2 concentration varies to a great degree. When olivine particles re-circulate from the riser and returns in a counter-current mode from the upper loop-seal into the gasifier, a part of the product gas can be immediately oxidized with a rapid oxidation reaction close to the gasifier inlet. Thus, the drop of the H_2 concentration from the BFB to the DFB mode can be well-explained by the olivine bed material as an oxygen carrier from the riser to the gasifier and by some introduction of air from the upper pressure lock. The H_2 concentration increases from the DFB mode without in situ reformer to the DFB mode with in situ reformer and increases further in DFB mode with in situ reformer filled with Ni-pellets. This can be attributed to the effective steam reforming of hydrocarbons in the reformer.

Corresponding to the H_2 concentration increase seen in Fig. 13, the tar content in the syngas decreases significantly for the mode with the in situ reformer filled with Ni pellets. Ni catalyst is highly effective in reforming of hydrocarbons such as tars and CH_4, and able to dramatically decrease the tar content and increase the hydrogen content in the syngas from biomass fluidised bed gasification [30–32].

4 Conclusion

An internal reformer is developed for in situ catalytic reforming of tar and methane in DFBGs. The study on the internal reformer operated with and without Ni-catalytic pellets was carried out by evaluation of the syngas composition and tar/CH_4 content. It can be concluded that the reformer with Ni-catalytic pellets clearly results in higher H_2 content together with lower CH_4 and tar contents in the syngas than the reformer without Ni-catalytic pellets. The gravimetric tar content decreases down to 5 g/m^3 and the CH_4 content down below 6 % in the syngas. These results suggest a strong endothermic reaction of steam reforming of hydrocarbons, such as tars and CH_4, using Ni-catalytic pellets in an in situ reformer placed in the hottest region of the gasifier. The novel design in the MIUN gasifier increases the gasification efficiency, suppresses the tar generation and upgrades the syngas quality.

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Glossary

BFB	Bubbling fluidised bed
CFB	Circulating fluidised bed
CLC	Chemical looping combustion
DFB	Dual fluidised bed
DFBG	Dual fluidised bed gasifier
GHG	Greenhouse gas
MIUN	Mid Sweden University
WGS	Water-gas-shift

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