



Principles of a novel multistage circulating fluidized bed reactor for biomass gasification

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Abstract

In this paper a novel multistage circulating fluidized bed reactor has been introduced. The riser of this multistage circulating fluidized bed consists of several segments (seven in the base-case design) in series each built-up out of two opposite cones. Due to the specific shape, a fluidized bed arises in the bottom cone of each riser segment. Back-mixing of gas and solids between the segments is prevented effectively. The absence of back-mixing combined with the enlarged solids residence time in each segment (each segment is a fluidized bed) creates the opportunity to operate, spatially divided, separate process steps in a single reactor. The benefit of a concept in which different processes are carried out in separate segments of the same reactor has been demonstrated for the specific case of biomass gasification. In the novel reactor it is possible to create oxidation segments in which O₂ reacts exclusively with char (carbon). This results in an increased carbon conversion and consequently improved gasification efficiency.

Creating an exclusive char combustion zone, aimed at improving both the carbon conversion and the thermal efficiency, has also been applied successfully in a conventional CFB biomass gasifier (ECN's CFB \approx 100 kg wood/h) by building a flow restriction in the riser between the primary air nozzles and the biomass feed point.

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1. Introduction

Sustainable sources of energy are bound to play an important role in the total energy production system in the near future. Biomass, which can be processed by various conversion methods like combustion, pyrolysis and gasification, can contribute as a CO₂ neutral and renewable source of energy.

In this paper a multistage reactor will be presented, which has been designed especially to overcome a number of problems usually encountered in circulating fluidized bed gasification of biomass. Circulating fluidized beds have been used for several processes, e.g. fluid catalytic cracking, Fischer–Tropsch synthesis, etc. Despite the many advantages like the high throughput per unit reactor volume and a good heat control, the overall gas–solids contacting efficiency (e.g. Venderbosch, Prins, & van Swaaij, 1999) and the state of axial and radial gas mixing (e.g. Kersten, Moonen, Prins, & van Swaaij, 2001) are not always optimal. Also the coupling

between the solids circulation rate, the gas velocity (profile) and the solids hold-up does not always provide well balanced reactor conditions. The present concept may create favorable reactor conditions that cannot be realized otherwise, e.g. (i) recycling of solids, while maintaining a high solids hold-up in the reactor, (ii) overall plug flow behavior for the gas and solids flow, (iii) no back mixing of gas and solids, (iv) the opportunity to combine separate processes, spatially divided, in a single reactor.

First the concept of the novel reactor together with its design aspects will be explained. This is followed by a discussion on the benefits of this concept with respect to the application of biomass gasification. After a description of the used experimental set-ups and the employed experimental methods, the results of a few cold-flow hydrodynamic characterization experiments are summarized. In a future paper the novel concept will be characterized in more detail. Results of measurements concerning the pressure drop (solids-hold-up), solids flux, gas ‘back’-mixing, gas residence time distribution (gas flow pattern), solids mixing, heat transfer and the influence of the riser geometry will then be presented. In the present paper the results of gasification

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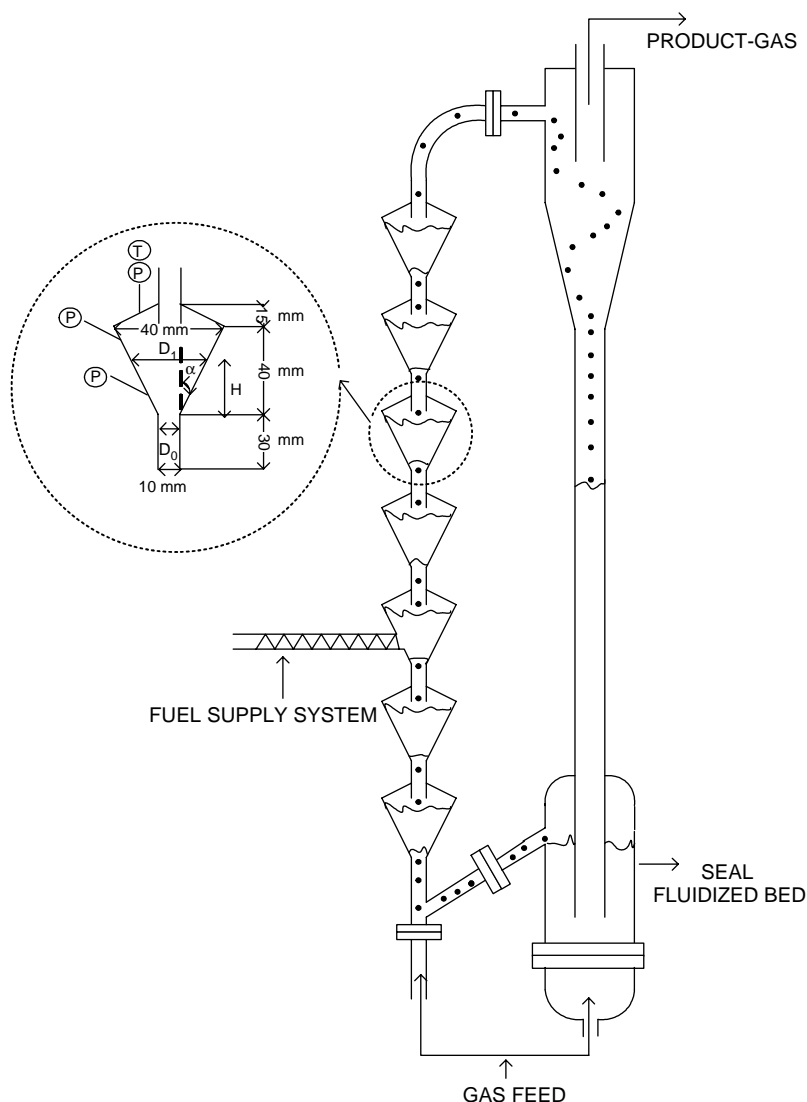


Fig. 1. Schematic representation of the novel multistage fluidized bed for biomass gasification.

experiments carried out in the novel reactor are reported as well.

2. The novel reactor

The novel reactor is a riser built-up of several segments, each being constructed from two opposite cones welded together (see Fig. 1). Tubes with a diameter equal to the base diameter (D_0) of the cones interconnect the segments. In fact the riser reactor has now a continuously varying cross-sectional area, which simultaneously allows the formation of a fluidized bed in the bottom cone of every segment, and the upward transport of solids through the tubes between them. It is important to note that the new reactor is a co-current multistage fluidized bed. The precise nature of the fluid beds (bubbling, turbulent or even spouting) depends of course on the applied gas flow rate through the

reactor, the properties of the solids and the geometry (cone angle, α) of the reactor segments. The gas–solids stream out of the last segment is separated by a cyclone: the solids are fed back to the riser whereas the gas leaves the system. This enables recycling of solids, while maintaining a high solids hold-up (FB in the bottom cone of each segment) in the reactor. In the base-case design the solids are recycled to the riser by means of a downcomer/seal fluidized bed system (see Fig. 1). Recycling of solids can be applied for instance after catalyst regeneration or heat exchange.

When the segments are operated above incipient fluidization it is possible to have “partially” fluidized beds present in the bottom cone of each segment of the riser without using any gas distribution plates, because for this condition the friction pressure drop of the gas balances the weight of the solids. By integrating Ergun’s (Ergun, 1952) equation, the incipient fluidization velocity of a conical fluidized bed can be calculated by assuming that the distribution of gas at

Table 1
Bed particles and their calculated gas-velocity characteristics

	d_p (μm)	GI	u_{mf} (m/s)	$u_{mf,cone}$ (m/s) ^a	u_t (m/s) ^b	u_{ms} (m/s) ^{c,a}	H_{jet} (cm) { $u_t(D_0) = 2.5u_t$ } ^d
Sand I	96	2.13	0.006	0.043	0.56	1.01–4.84	1.2–8.1
Sand II	182	2.40	0.021 (0.022 ^e)	0.155	1.16	1.20–4.82	1.1–7.9
Sand III	242	1.86	0.038 (0.037 ^e)	0.27	1.61	1.16–4.80	1.2–7.8
Sand IV	475	n/a	0.138	0.96	3.47	0.79–4.83	1.2–7.8

^aIt was assumed that $D_1 = D_{max}$.

^bThe drag coefficient was taken from Perry (1950).

^cThe range was calculated on basis of the relations (6) given in the paper of Olazar, San José, Aguayo, Arandes, and Bilbao (1992).

^dThe range was calculated on basis of the relations (5) given in the paper of Massimilla (1985).

^eExperimental values obtained in a normal fluidized bed.

All calculations were done for $T = 25^\circ\text{C}$ and $p = 1$ bar.

For all sand types, ρ was approximately 2600 kg/m^3 .

every cross section is uniform:

$$B \left(\frac{D_0}{D_1} \right) u_{mf,cone}^2 + A u_{mf,cone} - (1 - \varepsilon_{mf})(\rho_s - \rho_f)g \times \frac{D_1^2 + D_1 D_0 + D_0^2}{3D_0^2} = 0 \quad (1)$$

with

$$D_1 = D_0 + 2H \tan(\alpha).$$

Here $u_{mf,cone}$ is defined for the cross-sectional area at the cone base diameter D_0 , D_1 is the cone diameter at the fluid bed solids level in the bottom cone. Due to the increasing cross-sectional area and consequently decreasing linear gas velocity, the gas velocity at D_0 for which the conical bed is at incipient fluidization ($u_{mf,cone}$) can exceed many times u_{mf} . For coarse particles and large cone angles the incipient fluidization velocity at D_0 ($u_{mf,cone}$) can even be higher than the terminal velocity. Note that at $u_{mf,cone}$ the bed is not completely fluidized (only the weight of the solids is balanced by the gas pressure drop). Obviously for complete fluidization ($u_{mf,compl}$) the following equation holds:

$$u_{mf,compl} = u_{mf} \left(\frac{D_1}{D_0} \right)^2. \quad (2)$$

In order to transport particles between two segments the gas velocity in the interconnecting tubes (D_0) must exceed the terminal velocity of the particles. The local gas velocity at top of the fluidized bed in a bottom cone does not necessarily need to be higher than the terminal velocity to enable transport of solids upwards to the interconnecting tube. It is sufficient when this fluidized bed is in the bubbling, turbulent or spouted regime. In fact, the presence of splash or fountain zones above the various fluidized beds is characteristic for the operation of the novel reactor. The splash/fountain zone is always located (partly) in the top cone of each segment. From that zone the particles are entrained and elutriated by the gas flow which is accelerated by a factor $(D_{max}/D_0)^2$. To amplify the gas acceleration the interconnecting tube partially enters the top cone region (see Fig. 1). In the base

case design of Fig. 1, (D_{max}/D_0) equals 4 and the ratio between the volumes of the bottom and top cone equals 4/1.5. Table 1 gives the minimum fluidization velocities of the base case segments ($u_{mf,cone}$) for the bed materials used in this study.

Like mentioned before, the precise nature of the fluidized bed in the bottom cone of a segment depends, among others, on the applied gas flow rate. Correlations, originally derived for spouted beds, conical fluidized beds (Olazar et al., 1992) and for jet streams in normal fluidized beds (Massimilla, 1985), could be used to predict the regime of this bed. However, the inaccuracy of such correlations is far too high to yield reliable design information. To illustrate this, the predicted range of the minimum spouting velocity and the maximum jet dept is listed in Table 1. The correlations are applied for the solid materials and typical gas flows used in the base case design. In addition, it should be noticed that the system described here does not really resemble any comparable system (e.g. spouted beds): in this case there is a significant solids flux. All this clearly leaves the regime of the fluidized bed in the bottom cone open for further research.

Due to the high gas and solids velocity at the entrance and exit, back-mixing of gas and solids to lower segments is prevented completely.

Overall as well as per segment, the ratio between the solids and gas residence time is much higher than for a normal CFB. The combination of an increased solids residence time in one segment combined with the absence of back-mixing between segments creates the opportunity to perform different processes (e.g. gas–solids reactions, pyrolysis of biomass), spatially divided, in a single reactor. As a result of the staging, it is expected that the gas and solids flow closely approach overall a plug flow pattern. Plug flow combined with renewed contacting in each segment (staging) may provide better gas–solids contacting than conventional BFB/CFB reactors if properly scaled with the gas residence time. Just like in a normal fluidized bed, the degree of solids and gas mixing in each segment can be controlled by the gas velocity.

3. The application of biomass gasification

Biomass gasification is a conversion process, which transforms a solid biomass fuel into a gaseous energy carrier (CO, CO₂, CH₄, C₂₊, H₂, H₂O). The main reaction steps in biomass gasification are:

- heating and pyrolysis of the biomass (biomass is converted to gas, char and primary tar),
- cracking of primary tar to gasses and secondary and ternary tars,
- cracking of secondary and ternary tars,
- heterogeneous gasification reactions of the char formed during pyrolysis and homogeneous gas-phase reactions (e.g. the shift reaction),
- combustion of char formed during pyrolysis and oxidation of combustible gases.

It is common practice to characterize the gasification process by the equivalence ratio (λ), which is the ratio between the actual amount of oxygen supplied to system and the amount of oxygen that is needed for complete combustion of the fuel. At present a major problem of conventional CFB gasification technology is the low thermal efficiency due to incomplete carbon conversion. This is caused by a strong competition between char combustion and combustion of volatile components (H₂, CO, tars, etc.) in the oxidation zone of such a gasifier (Kersten, Prins, van der Drift, & van Swaaij, 2002). As a result of this competition, the carbon conversion is incomplete in practice, especially at low equivalence ratios. In the novel reactor concept, an exclusive char combustion zone can be created in the two bottom segments of the riser if the biomass is fed to the third segment (see Fig. 1). Combustible gaseous components cannot enter the oxidation segments due to the absence of back-mixing. When, in addition, the char hold-up in the two bottom segments is high enough to ensure that oxygen does not break through to the feed segment, exclusive reaction of O₂ with char is ensured. Char enters the oxidation segments together with the other solids (inert and, possibly, catalyst) via the re-circulation loop. Like in regular CFB gasifiers the upward solids flux enables the transport of energy from the oxidation zone to the pyrolysis and tar cracking segments (discussed hereafter).

The high solids residence time in the feed segment may create the possibility to achieve almost complete conversion of the biomass devolatilization process in this segment. Another major problem in biomass gasification technology is the too high concentrations of secondary and ternary tar in the product gas for direct use in prime movers. Tars can be cracked catalytically at typical CFB gasification temperatures (say 750–1000°C) over dolomite or Ni-catalysts (e.g. Ekström, Lindman & Pettersson, 1982). An elegant solution may be to use the

catalytic activity of pyrolysis char formed in situ to crack the secondary and ternary tars (see also Ekström, et al., 1982). While considering the cracking of tars, the advantage of the presented multistage fluidized bed compared to a CFB riser is the high solids (catalysts) hold-up in each riser segment. In the proposed system, tar cracking is expected to proceed mainly in segments 4–7. Compared to both a BFB and a CFB the expected advantage is, that due to the specific properties of each segment and the staging of these segments, overall, the solids and the gas flow approaches plug flow with the accompanying improved gas–solids contacting efficiency. Especially when a high degree of conversion is required of a gas phase component (tar cracking) the gas–solids contacting efficiency is crucial.

4. Experimental set-ups

Hydrodynamic characterization of the novel multistage fluidized bed has been carried out in a cold-flow set-up made of glass. This set-up will be described in more detail in a future paper. Biomass gasification experiments were carried out in the reactor presented in Fig. 1. The reactor is made from stainless steel. Hereafter the flow-sheet of the mini-plant is described. The biomass is fed from a hopper via a screw to the third element. This screw is water cooled in order to prevent biomass devolatilization and consequently char and tar formation in the screw. The average biomass feed rate is determined by weighing the hopper content before and after an experiment. Willow- and pinewood were used as feedstock (both approximately: 10 wt% moisture, 1.5 wt% ashes, $d_p \approx 1.5$ mm). Air and/or nitrogen are fed to both the riser and the seal fluidized bed by means of mass flow controllers. Because of the small scale of the set-up (100 g/h biomass) it is impossible to reach auto thermal operation. Therefore the reactor is installed into an oven (4 kW), which allows an independent temperature control. Inside the reactor the temperature and the pressure are measured in each segment of the riser and at several other positions. The reactor exit is connected via a secondary cyclone to the condenser. Up to the condenser, the tubing (including the 2nd cyclone) is kept at 400°C by trace heating to prevent condensation of tars and water. After the condenser ($\pm 5^\circ\text{C}$) the product is purged into the main ventilation system of the laboratory. A sub-stream is passed through a gas analysis section. Before entering this section, the gas passes through a dust-filter and an additional cooler to condition the gas to the requirements of the analyzers. The following components could be detected: H₂ (thermal conductivity), O₂ (para-magnetic), CO (IR), CO₂ (IR) and CH₄ (IR). Occasionally, also a micro-GC was used to measure hydrocarbons up to xylene. The outgoing dry gas flow rate is determined on basis of the nitrogen balance. This method turned out to be more accurate than measurement of the flows.

Table 2

Total gas yields (exp. nos. 1 and 2), and the gas yields of the combustion zone only (exp. nos. 1-2S and 2-2S) for wood gasification in the novel reactor

Exp. no.	Temp. (°C)	λ	τ^a (s)		CO	CO ₂	CH ₄	C ₂ H ₄	C ₆ H ₆	C ^b		H ₂	H ₂ O
1	740	0.0	0.86	Y_t	0.33	0.19	0.08	0.05	0.04	0.12	Y_t	0.019	0.29 ^c
				Y_c	0.31	0.11	0.13	0.10	0.08	0.26	0.23 ^d		
1-2S	740	0.0	0.24	Y_t	0.00	0.00	0.00	0.00	0.00	n/a	Y_t	0.00	n/a
2	740	0.19	0.86	Y_t	0.33	0.50	0.08	0.05	0.04	0.035	Y_t	0.019	0.14 ^a
				Y_c	0.30	0.31	0.13	0.09	0.08	0.08	0.21 ^b		
2-2S	740	0.19	0.24	Y_t	0.01	0.33	0.00	0.00	0.00	n/a	Y_t	0.00	0.19 ^c
				Y_0	0.04	0.95				n/a			

^aCalculated by dividing the available gas volume (Δp determines the voidage) by the average volumetric flow rate.^bCalculated as 1 minus the measured carbon containing components in the gas.^cCalculated with the hydrogen balance.^dCalculated with the oxygen balance.^eCollected in condenser.

5. Experimental results

5.1. Cold-flow characterization

A few results of the cold-flow characterization are summarized hereafter. More details will be published in the near future.

- The reactor showed no start-up problems. If there is enough solid material present in the system, a steady-state situation is reached within a few seconds after the gas flow to the riser and the seal fluidized bed are turned on. In this steady state, a fluidized bed is clearly visible in the bottom cone of each segment.
- Besides the total amount of inert solids in the system, the gas velocity through the riser and the seal fluidized bed are the most important operation variables; they control the solids flux, the solids hold-up and heat transfer in each segment, and the degree of mixing in a segment.
- Back-mixing of gas and solids between the segments is prevented effectively. This has been demonstrated by gas-tracer experiments (CO₂ and He).
- Gas-phase RTD measurements showed that: (i) there is no tailing, (ii) the mean gas residence time calculated on the basis of the measured RTD-curves is in very good agreement with the actual holding time (τ) of the riser and (iii) the overall gas-phase hydrodynamics can be described with a CISTR cascade model, in which the number of CISTRs is almost the same as the real number of segments of the riser.

5.2. Results of biomass gasification

The laboratory gasifier (nominal 100 g wet biomass/h) could be operated without any serious problems. A standard experiment is started at an equivalence ratio of zero (N₂ is

fed to both the riser and the seal fluidized bed) for approximately 30 min to build-up a realistic char concentration in the reactor. Then air is added to the reactor to achieve the desired equivalence ratio. Finally, the experiment is continued for 1 or 2 h at the selected operation temperature. Due to the cooled screw, which is connected to the feed segment, the temperature in this particular segment is about 15°C lower than in the other segments. The temperature in the feed segment is chosen to be the process temperature.

Back-mixing of vapors from the biomass feed (3rd) segment to the two bottom combustion segments has been examined by measuring the gas composition in the 2nd segment during gasification experiments at an equivalence ratio of zero (only nitrogen added). No product gases have been detected in the second segment during these tests (see Table 2, exp. 1-2S), which means that back mixing of biomass or combustible vapors in these segments is prevented effectively. However, oxygen can still contact these combustible vapors if it slips through the combustion segments. That is possible if there is not enough char available in these segments or in case the residence time is too short to reach complete conversion of oxygen. This feature of the new reactor is demonstrated from the measured mass balance over the oxidation segments during gasification conditions (see Table 2, exp. 2-2S for a typical result). The closure of the mass balance of the oxidation segments is normally within 2%, and shows very clearly that the oxygen added to the reactor reacts only with char in these segments to produce CO₂ and CO (see Table 2, exp. 2-2S). Summarizing, by feeding air in the first segment and biomass in the third segment from the bottom, a separate combustion zone was created in the two bottom segments. Here the oxygen is consumed exclusively and completely by reaction with the recycled char introduced underneath the first segment.

In Fig. 2 the results of gasification experiments in the novel multistage fluidized bed are compared with results of

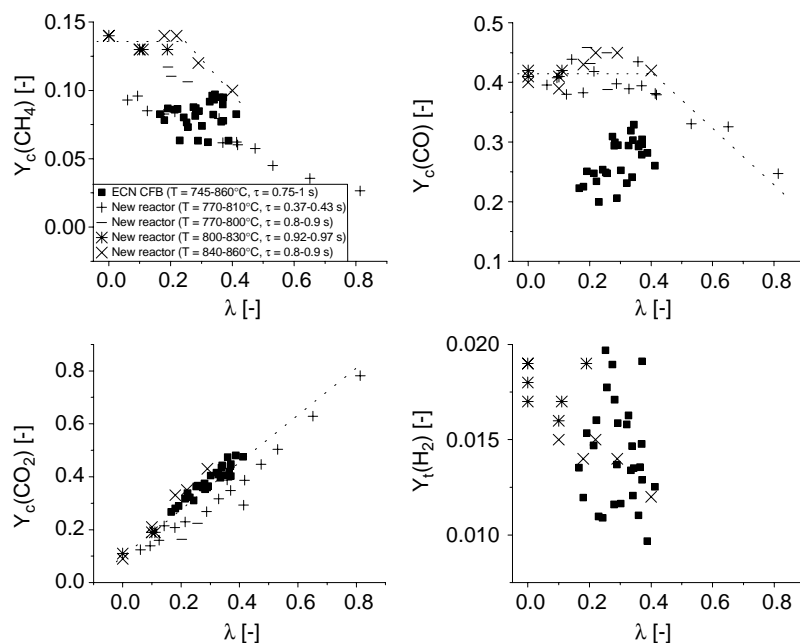


Fig. 2. Measured gas yields versus the applied equivalence ratio. Results of the novel reactor are compared with results of ECN's CFB pilot gasifier.

gasification tests in ECN's pilot CFB gasifier (100 kg/h \simeq 500 kW_{th}). The results show that in the novel multistage fluid bed reactor the hydrocarbon (CH₄, C₂H₄, etc.) yield increases for higher gas residence times (see Fig. 2: CH₄). A higher conversion of the primary tars at larger residence times may explain this. Fig. 2 shows clearly that especially the yield of CO, but also the yield of CH₄ in the new reactor is higher than in ECN's CFB biomass gasifier at comparable temperatures and holding times (τ). The CO₂ yield is approximately the same. Overall, for low equivalence ratios (up to 0.30), the carbon conversion is higher than observed in ECN's CFB gasifier, while for higher equivalence ratios (say $\lambda > 0.35$) the carbon conversion of both reactors is nearly complete. The hydrogen yield obtained in the small novel reactor is in good agreement with the hydrogen yield of the large ECN-CFB gasifier. However, it should be noticed that the scatter in the hydrogen balance is always large (see Fig. 2).

In the lab-scale multistage CFB, the yields of CH₄ and CO remain approximately constant at increasing the equivalence ratio up to a value of 0.25 and 0.4, respectively (see Fig. 2). This again proves that in the bottom two segments of the multistage CFB a separate oxidation zone is created where at least for low equivalence ratios (say up to 0.3) enough char is available for complete conversion of oxygen. If oxygen would slip through the oxidation segments, the yields of CO and CH₄ would have decreased with increasing λ . An important conclusion is that, if a separate char combustion zone can be realized in commercial large-scale (hardly any heat-loss) gasifiers, almost complete carbon conversion and a thermal efficiency over 80% can be achieved at a process temperature of around 800°C.

6. Conclusion and discussion

In this paper a novel multistage circulating fluidized bed reactor has been introduced. The riser consists of several segments in series, each built-up of two opposite cones. Due to its specific shape, a fluidized bed arises in the bottom cone of each segment and back-mixing of gas and solids between the segments is prevented effectively. The concept of several fluidized beds operated co-currently in series results in a ratio between the solids residence and the gas residence time, that is much higher than for a normal CFB. The absence of back-mixing and the large solids residence time per segment offers the opportunity to combine separate processes (e.g. gas–solids reactions), spatially divided, in a single reactor. This has been demonstrated for the application of biomass gasification. It has turned out that when feeding the biomass in the 3rd segment of the multistage fluidized bed, an oxidation zone in the two bottom segments is created in which enough char is present to convert all incoming oxygen to CO and CO₂ up to large-scale equivalence ratios (say $\lambda = 0.25$). This results in an increase of the carbon conversion and consequently improved gasification efficiency. Creating an exclusive char combustion zone by staging has also been tested in ECN's pilot CFB facility. A flow restriction was constructed below the biomass feed point, which is located one meter above the primary air nozzles (see Fig. 3). In this way, when enough char is present, an exclusive char combustion is created. At a typical gasification equivalence ratio of 0.25 the carbon conversion increased from 87% to 96% and the thermal efficiency from 65% to 70% on average (Van der Drift, Van der Meijden, & Strating-Ytsma, 2002). Scaling up these data to a commercial large-scale

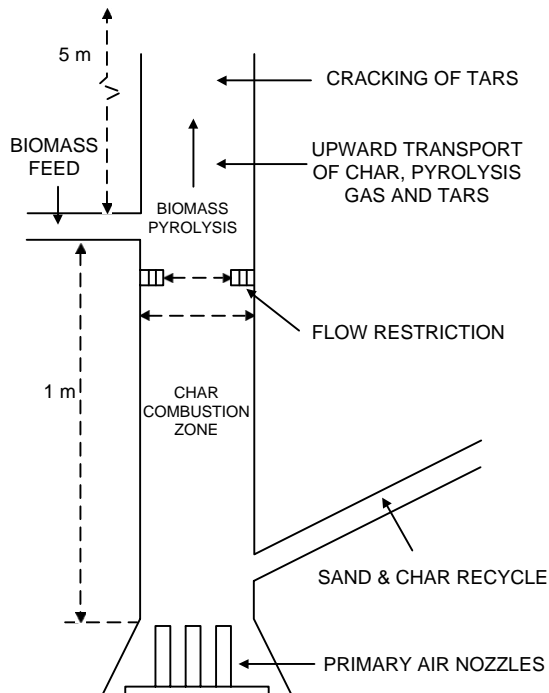


Fig. 3. Sketch of the modified bottom part in ECN's pilot-plant CFB gasifier. Locations where the sub-processes of gasification (char combustion, pyrolysis, tar cracking) are supposed to take place are indicated.

gasifier with almost no heat loss would lead to a thermal efficiency of 80% with nearly complete carbon conversion.

Notation

A	constant in Ergun's equation, $\text{kg}/(\text{m}^3 \text{ s})$
B	constant in Ergun's equation, kg/m^4
D_0	inlet (smallest) diameter ($=0.01 \text{ m}$ in the base-case) of the bottom cone, m
D_1	diameter at the solids level in the bottom cone, m
D_{max}	maximum diameter ($=0.04 \text{ m}$ in the base-case), m
d_p	particle size, μm or mm
$d_{p,\text{max}}$	maximum particle size of the size distribution, μm or mm
g	gravitational acceleration constant, m/s^2
GI	gradation index: fraction of particles smaller than $0.9d_{p,\text{max}}$ divided by the fraction of particles smaller than $0.1d_{p,\text{max}}$, (dimensionless)
G_s	solids flux, $\text{kg}/(\text{m}^2 \cdot \text{s})$
H	height of solids in the bottom cone, m
H_{jet}	jet penetration height in a fluidized bed, m or cm
p	pressure, bar or Pa
T	temperature, $^\circ\text{C}$ or K
u_{mf}	minimum fluidization velocity, m/s
$u_{mf,\text{compl}}$	gas velocity at D_0 above which the conical FB is completely fluidized (defined in Eq. (2)), m/s
$u_{mf,\text{cone}}$	gas velocity at D_0 for which the conical FB is at minimum fluidization (defined in Eq. (1)), m/s

u_{ms}	minimum spouting velocity, m/s
u_r	gas velocity through the riser (at D_0), m/s
u_t	terminal velocity, m/s
Y_c	carbon yield: gram C (carbon) in component divided by gram C in biomass, (dimensionless)
Y_o	oxygen yield: gram O in component divided by gram O in air, (dimensionless)
Y_t	total yield: gram component divided by gram wet biomass, (dimensionless)

Greek letters

α	cone angle, (dimensionless)
ε_{mf}	voidage at minimum fluidization, (dimensionless)
λ	equivalence ratio, (dimensionless)
ρ_f	gas density, kg/m^3
ρ_s	solids density, kg/m^3
τ	gas holding time (available gas volume divided by the gas flow rate), s

Abbreviations

BFB	bubbling fluidized bed
CFB	circulating fluidized bed
FB	fluidized bed

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