Pressurized Entrained Flow Gasification of Pulverized Biomass – Experiences from Pilot Scale Operation

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One of the goals in the national energy strategy of Sweden is that the vehicle fleet should be independent of fossil fuels by 2030. To reach that goal and to domestically secure for supply of alternative fuels, one of the suggested routes is methanol production from forest residues via pressurized and oxygen blown entrained flow gasification. In this context, ongoing industrial research in a 1 MW_{th} gasification pilot plant is carried out at SP Energy Technology Center (SP ETC) in Piteå, Sweden. The plant is operated with pulverized or liquid fuels at process pressures up to 10 bar and this work summarizes the experiences from over 600 hours of operation with forest based biomass fuels. This paper covers results from thorough process characterization as well as results from extractive samplings of both permanent gases and particulate matter (soot) from inside the hot gasifier. Furthermore, the challenges with pressurized entrained flow gasification of pulverized biomass are discussed. During the characterization work, four of the most important process parameters (i.e. oxygen stoichiometric ratio (λ), fuel load, process pressure and fuel particle size distribution) were varied with the purpose of studying the effect on the process performance and the resulting syngas quality. The experimental results showed that the maximum cold gas efficiency (CGE) based on all combustible species in the syngas was 75% (at λ=0.30), whereas the corresponding value based only on CO and H_2 (with respect to further MeOH synthesis from the syngas) was 70% (at λ=0.35). As expected, the pilot experiments showed that both the soot yield and soot particle size was reduced by increasing λ. One of the additional conclusions from this work was that; minimizing heat losses from the gasifier is of utmost importance to optimize the process performance regarding energy efficiency (i.e. CGE). Therefore, a well-insulated refractory lined gasifier is the primary alternative in regards to reactor design to maximize the CGE. Future development of the PEBG process should focus on identifying suitable hot-phase refractory, that exhibit long life-time and can sustain the alkali-rich biomass ash under gasification conditions. In addition to this, the remaining issue around how to improve the slag flow from the reactor, by additives or fuel mixing, should be investigated.

1. Introduction

One of the goals in the national energy strategy of Sweden is that the vehicle fleet should be independent of fossil fuels by 2030 (Swedish Government, 2009). Pressurized, oxygen blown, entrained flow gasification of forest residues followed by methanol production is one of the suggested routes for the production of synthetic motor fuels that could help reach this goal (Börjesson and Ahlgren, 2012). One of the benefits with entrained flow gasification, compared to other gasification technologies, is that a syngas with high quality is generated (Higman and van der Burgt, 2008). The high syngas quality is necessary for the subsequent synthesis to biofuels, where impurities otherwise can cause problems (Woolcock and Brown, 2013). In this context, ongoing industrial research in a 1 MW_{th} gasification pilot plant is carried out at SP Energy Technology Center (SP ETC) in Piteå, Sweden. The gasifier can be operated both with pulverized and liquid fuels at process pressures up to 10 bar. This work summarizes the experiences from over 600 hours of operation with forest based biomass fuels. The paper covers results from thorough process characterization as well as results from extractive samplings of both permanent gases and particulate matter (soot) from inside the hot gasifier. Furthermore, the challenges with pressurized entrained flow gasification of pulverized biomass are discussed.

2. Method

The Pressurized Entrained flow Biomass Gasification pilot plant (PEBG) was designed to operate in slagging mode with process temperatures up to 1600 °C. A detailed description of the plant can be found elsewhere (Weiland et al., 2013) and therefore only a brief description is given herein. The PEBG gasifier consists of a ceramic (mullite based, Vibron 160H) lined reactor (0.52 m inner diameter and approximately 2 m long) followed by a water sprayed quench for syngas cooling and smelt/particle separation. The burner where the pulverized fuel and oxidant (O_2) were injected was installed in the top of the reactor. In the case of pyrolysis oil gasification, the feeding system for solid fuels was replaced with an oil feeding system and a liquid fuel burner nozzle. The raw syngas and other products from the reactor (i.e. gases, particulates and ash/char) were cooled to temperatures below 100 °C by water sprays in the quench. A small slip stream of the cooled syngas was continuously sampled and analyzed by a micro GC for the concentrations of the syngas species; CO, H_2, CO_2, CH_4, C_2H_4 and C_2H_2. Additionally, a water cooled sampling probe with N_2-dilution was used in a number of experiments to sample gas and particulates from inside the hot gasification zone. Finally, the excess syngas was flared to eliminate any risks connected to the syngas.

From the PEBG-project start-up, the plant has been operated over 600 h with different biomass fuels such as e.g. stem wood powder from spruce and pine (e.g. Weiland et al, 2013 and Weiland et al., 2015), bark and bark/peat mixtures, pyrolysis oil from pine and straw (Leijenhorst et al, 2015), lignin residues from wheat straw (Öhrman et al., 2013) as well as forest residues pretreated by torrefaction (Weiland et al., 2014). Most of the work has been done within the PEBG-project in close collaboration between SP ETC, Luleå University of Technology and Biogreen. In addition, the PEBG plant has been used in cooperation with other research partners (e.g. BTG, Enschede, The Netherlands; CEA, Grenoble, France; SINTEF, Trondheim, Norway; BioEndev, Umeå, Sweden and National Technology University of Athens, Greece) to study the gasification performance of alternative feedstock at different conditions or to study the raw syngas cleaning.

Some of the most important experimental results so far come from a thorough process characterization that was conducted with pulverized stem wood fuel. In that study, four of the most important process parameters (i.e. oxygen stoichiometric ratio (λ), fuel load, process pressure and fuel particle size distribution) were varied in order to study the effect on the process performance and the syngas quality (Weiland et al., 2015). In addition to the scientific knowledge, the experiments have also resulted in extensive practical experience regarding the gasifier operation.

3. Results and discussion

3.1 Characterization of the process performance

The most important process variable during gasification is the oxygen stoichiometric ratio, λ. This parameter strongly controls the process temperature. Increasing λ means increasing the proportion of oxygen fed to the gasifier and, thus, more heat is released by the combustion reactions. As an example, the measured process temperature at 600 kW fuel load and $\lambda=0.30$ was approximately 1180 °C. Additional increase of λ in steps of 0.1 resulted in temperature increasing approximately 150-200 °C per step. The fuel conversion was found to be significantly controlled by the process temperature. Additionally, the temperature was shown to have a dominating influence on the CH_4 yield, Figure 1, where several different fuels and operating conditions are presented. Beside the standard stem wood reference fuel, a spruce reference fuel and the corresponding torrefied spruce fuel is presented in Figure 1. In addition to this, a beech wood fuel with two different fuel powder particle size distributions were also gasified at varying operating conditions (2-7 barA). Regardless of the fuel type or the fuel pretreatment the yield of CH_4 seems to be clearly correlated to the process temperature. Interestingly, two adjacent points in the graph can originate from different stoichiometric conditions during gasification. This graph, therefore, indicates that the CH_4 yield was mainly governed by the process temperature and that the operating conditions (e.g. λ and fuel load) only were control parameters for the temperature.

Examples of experimental yields of the major syngas components (CO, H_2 and CO_2) and CH_4 are presented in Figure 2. The yields of CO and H_2 exhibited optima at $\lambda\sim0.42$ and $\lambda\sim0.35$, respectively. The syngas composition of H_2 and CO was such that the ratio H_2/CO was about 0.6. Therefore, the syngas requires shifting towards higher H_2 content if the syngas production is intended for methanol synthesis (should be ~2). The yield of CO_2 was increasing with λ because of the increased proportion of syngas combustion.

The cold gas efficiency (CGE) is a common measure for the gasification efficiency. It is defined as the ratio between the chemical energy in the cold syngas and the energy input from the corresponding fuel (based on the lower heating value). Depending on the purpose of the syngas generation, different measures of process efficiencies can be calculated. The CGE calculated from all combustible species in the syngas is in this case

denoted CGE_{power}. This is a representative measure if the syngas is intended for combustion in e.g. a gas turbine for power production, where all combustible species of the syngas can be used.
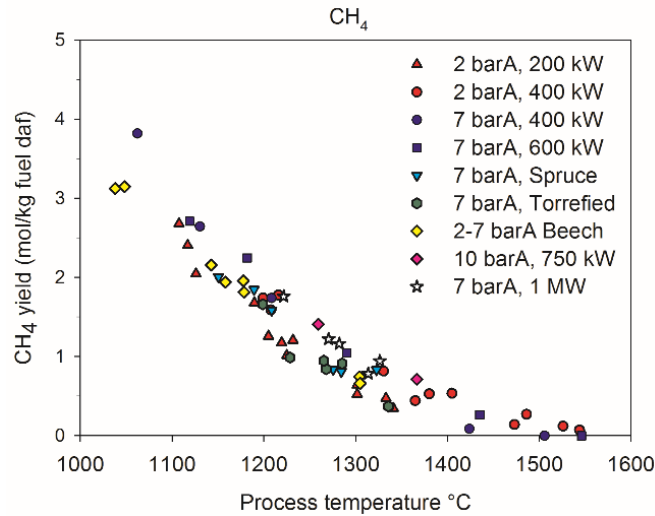

Figure 1 The yield of CH_4 (mol/kg fuel dry ash free) as a function of measured process temperature in the gasifier for several different operating conditions.
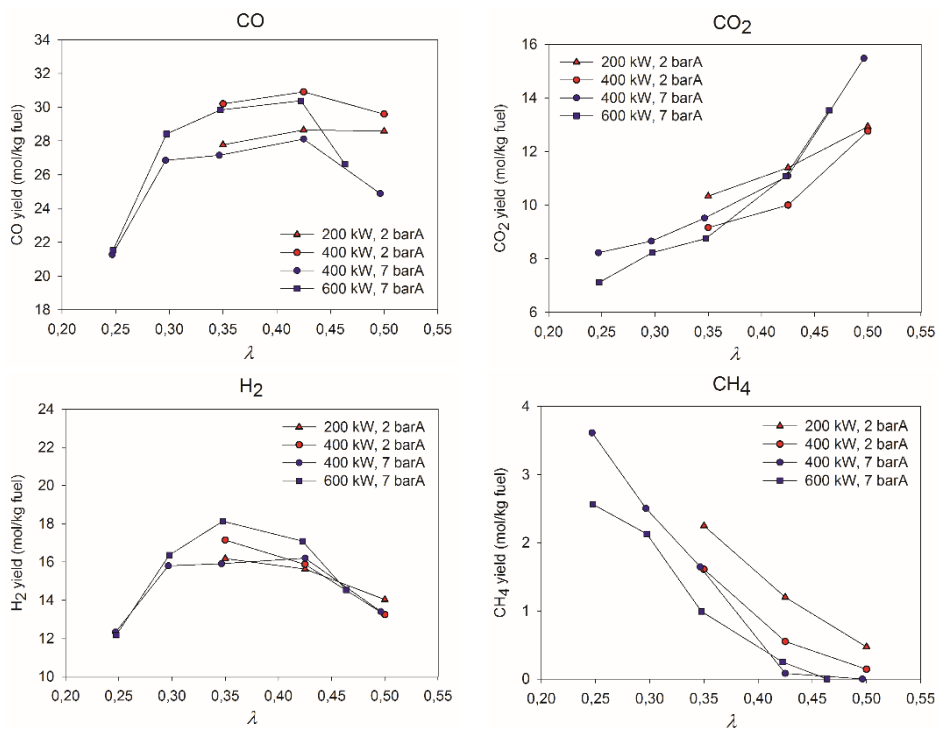

Figure 2 Examples of syngas yields of CO, H_2, CO_2 and CH_4 at different operating conditions.

The CGE_{fuel}, on the other hand, is a representative measure if the syngas is intended for catalytic synthesis of biofuels, where only the yields of CO and H_2 in the syngas should be maximized. The experimental CGEs can be found in Figure 3, where it can be seen that CGE_{power} reached a maximum of approximately 75 % (at $\lambda=0.30$). Lower λ resulted in poor fuel conversion and thereby a reduction in efficiency. Excessive combustion of the energetic syngas species was responsible for the CGE_{power} reduction at higher λ (above 0.30). The correlation between CGE_{fuel} and λ had a less pronounced optimum being rather flat around the maximum,

over a broad range of λ. This suggests that it is possible to operate the gasifier at an elevated λ without sacrificing too much of the CO and H_2 yields that constitutes the CGE$_{fuel}$. The benefit of operating the gasifier at an elevated λ is that it improves the syngas quality due to the conversion of CH_4 and other unwanted species (e.g. larger hydrocarbons and soot) because of the higher process temperature. Furthermore, an elevated λ can facilitate more effective slag removal at the reactor outlet due to the reduction in slag viscosity at higher temperature.
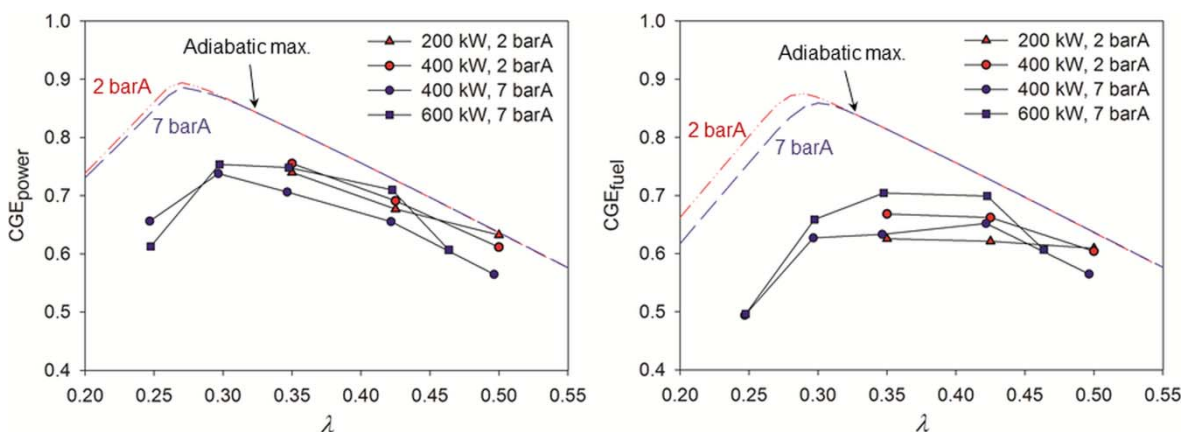

Figure 3 Cold gas efficiency at different operating conditions, CGE$_{power}$ (left) and CGE$_{fuel}$ (right).

3.2 Fuel feeding fluctuations

In a theoretical "perfect" gasifier, the fuel feeding and oxidant feeding rates are constant and without fluctuations. If the gasifier is well designed, this means that the point where complete gasification is reached lies slightly upstream the reactor outlet so that the syngas has time to equilibrate before leaving the gasifier. In the PEBG case, the oxygen feeding rate can be considered as constant (controlled by mass flow controller), whereas fluctuations can be expected in the fuel feed due to variations in (1) fuel powder bulk density, (2) fuel particle size distribution, (3) fuel composition and (4) limitations of the solid fuel feeding system itself. The point of complete fuel conversion is therefore expected to drift up and down in the gasifier as a result of these variations. For example the times for pyrolysis and char gasification are different for two differently sized fuel particles. A large fuel particle is expected to release pyrolysis products further down in the reactor compared to a small particle. During the pyrolysis stage, primary volatile compounds are formed by the separation of functional groups from the fuel structure. Subsequently, the primary volatiles decompose through secondary reactions to form e.g. CH_4, H_2, CO etc. (Warnatz et al., 2006). Fluctuations in for example the CH_4 concentration at the reactor probe position can therefore be considered as an indication of varying fuel particle size distribution.

The diluted reactor probe sample was sent to an FTIR instrument to monitor the syngas concentrations of CO, CO_2 and CH_4 at a sampling frequency of 5 Hz (1 Hz for Experiment 3, i.e. λ=0.41). Examples of the CH_4 concentration fluctuations are found in Figure 4 (left), where the CH_4 concentrations (mol-% dry N_2 free) for the three operating conditions are presented over a 200 s timespan. The concentration profiles show no clear signs of cyclical variation pattern. However, the fluctuations were relatively large (approximately ±0.3 mol-% dry N_2 free) compared to what is desirable for this type of process. This suggests that the fuel particle size distribution was not always constant during the timespan.

The O/C molar ratio in the sampled syngas (Figure 4, right) is considered as an indication of the stoichiometry fluctuations inside the gasifier. Since the O_2 feeding rate was constant, the local stoichiometric variations at the probe sampling position may be the result of fuel mass flow variations in combination with poor mixing inside the gasifier. The average O/C ratios for λ=0.41, λ=0.37 and λ=0.32 were approximately 1.32, 1.16 and 1.31, respectively. The surprisingly high O/C ratio at λ=0.32 was a result of the reduced carbon conversion compared to the other two experiments. It may be explained by much of the carbon was bound in the solid phase (soot and/or unconverted char) and that the gas phase, therefore, saw an unexpectedly high O/C molar ratio. However, the fluctuating O/C ratio suggests that also the fuel mass flow into the gasifier varied significantly over the timespan.
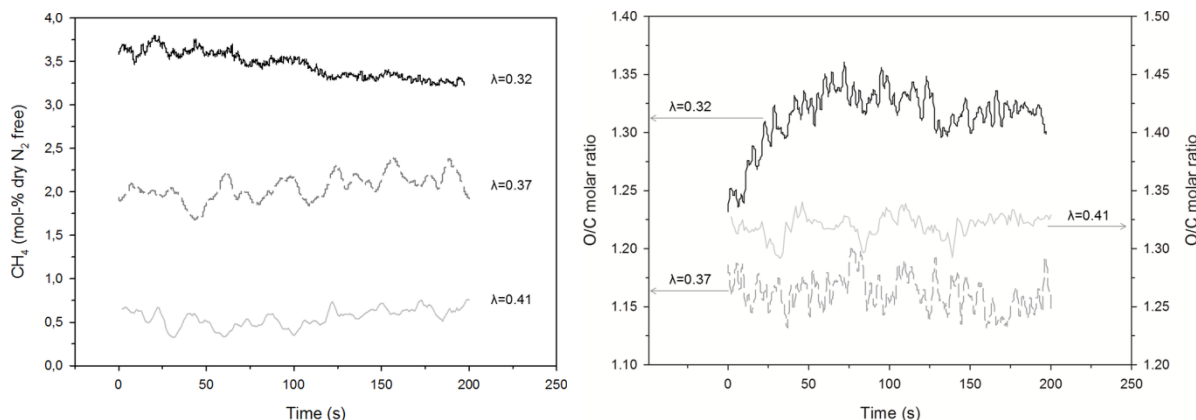
Figure 4 (left) Syngas CH₄ concentration (mol-% dry N₂ free) and (right) O/C molar ratio (note that the λ=0.41 experiment is on the right hand side ordinate).

3.3 Ash/slag interaction with mullite refractory

Experience showed that fuel ash adhered to the reactor wall where it reacted and dissolved the mullite based refractory (Vibron 160H). Increase in temperature decreased the melt viscosity resulting in a flowing slag, which resulted in loss of refractory material (Carlsson et al., 2014). It is, therefore, of outmost importance for the future to find refractory materials that can sustain the hot gasification environment in combination with the alkali rich biomass ashes. Moreover, the flowing slag blocked the reactor outlet at some occasions because of an increased slag viscosity, due to the lower temperature, near the outlet. To manage this issue, measures must be taken in order to avoid slag freezing at the bottom part of the reactor, e.g. better insulation from the quench or the possibility to inject O_2 close to the outlet in order to increase the temperature.

4. Conclusions

The 1 MW_{th} pilot-scale pressurized entrained flow gasification plant at SP ETC in Piteå, Sweden, has successfully been used for research purposes both internally at SP ETC, but also in a number of collaborative projects with other European partners. A thorough characterization of the process performance was conducted. It was found that the maximum yields of energetic gases in the syngas represented approximately 75 % of the energy content in the ingoing stem wood fuel. This value is expected to increase towards the theoretical maximum (89 %) in a commercial scale plant, where the proportion of the heat loss is smaller. It was concluded that a well-insulated refractory lined reactor is the preferred alternative regarding reactor design in order to optimize the process efficiency. Depending on the fuel ash melting temperature, it may be required to operate the gasifier at an elevated λ to ensure effective slag removal from the reactor. The experimental results showed that the H_2/CO ratio was only slightly altered in the attractive interval of operated λ. Another alternative may be the addition of a fluxing agent (or effective fuel mixing to obtain appropriate ash composition), which can decrease the ash melting temperature. A 100 °C decrease in the ash melting temperature would imply that the gasifier potentially can be operated at 0.05 units lower λ than without the additive (or fuel mixing). Consequently, the CGE would increase with approximately 2-6 %-units. There are, however, remaining challenges before this process can be commercialized; (1) suitable hot-phase refractory, that exhibit long life-time at the prevailing biomass gasification conditions, must be identified and tested under real conditions and (2) the fuel feeding fluctuations should be minimized in order to increase the process performance. Furthermore, efforts will be made to optimize the process efficiency by e.g. burner development, fuel milling efficiency and process control.

Acknowledgments

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